

Light-matter interaction is a fundamental phenomenon which is responsible, among others, for a large part of our ability to perceive the world around us. However, it is usually a weak one and either many photons and/or molecules/atoms are needed. Fortunately, the efficiency of this interaction can be enhanced by specially prepared structures, so-called cavities, which enhance light locally into e.g. hot spots. In those places interaction with matter is greatly amplified in a process called enhanced spontaneous emission. The strength of this interaction can, in fact, be amplified to such great values, that the two elements – light and matter – become to some extent indistinguishable.

When this occurs, the two states (the cavity mode and e.g. an electronic transition in matter) exchange energy between themselves with rates faster than any dissipation. In this strong coupling regime new so-called polariton modes arise which exhibit properties of both light and matter. This has made possible observation of unique phenomena such as photon-photon interactions: Bose-Einstein condensation of exciton polaritons and photon blockade. The strong coupling phenomenon has also garnered significant attention due to possibilities of modifying properties of matter simply by placing them in an appropriate optical resonator – one of such examples would be modified chemical rates for efficient chemistry/catalysis. To understand and predict these phenomena simple classical models and advanced quantum optical techniques are used, however, typically simplifications in the description of the material subpart are made in order to solve particular problems.

This project aims, on one hand, at lifting these limitations and to study strong coupling between molecular transitions or semiconductor excitons and plasmonic cavities using methods which ensure atomic scale resolution and account for electronic properties of the studied system. Using density functional theory, we aim to probe the feasibility of reaching single-molecule strong and ultra-strong coupling and proving that the energy landscape in such a plasmon-molecule strongly coupled system is indeed modified to the point of significantly affecting the molecule's chemistry. These electronic calculations will be accompanied by electromagnetic studies of novel extended nanostructures composed of bulk (multilayer) transition metal dichalcogenides (TMDCs). These 2D semiconductors (which can be stacked due to van der Waals forces) support excitons and due to a large background permittivity can form self-hybridized polaritons by coupling their own excitons and their own optical modes. These will provide a base for novel devices based on a strongly coupled light-matter system for efficient manipulation, detection, or harvesting of light as well as photocatalysis applications due to efficient modification of energy landscapes. The two methods will also be used jointly to provide accurate nonuniform electric excitation fields for time-resolved electronic studies or vice-versa with electronically accurate polarizabilities of nanoscale objects. The results obtained by these methods will serve as input and/or be subsequently interpreted with analytical treatments.

The outcomes of the project will further the development of strong light-matter coupling in nanoscale systems. On one hand it will provide new knowledge on self-hybridized excitonic optical cavities which could form the basis of easily tunable devices for energy harvesting, photodetection, light guiding or switching. The second novel aspect is the understanding at the atomic level how strong coupling with plasmonic nanocavities affects the energy landscape of molecules and how realistic it is to modify chemical reactivity at the single-molecule level or in a collective state (wherein many molecules couple coherently with the cavity).