Optical nonlinear properties for solids and nanostructures: theory and numerical simulations

A comprehensive understanding of the optical properties of solids is crucial for the improvement of non-linear materials and devices. It offers the opportunity to search for new materials with very specific properties. One particularly important process is **second harmonic generation**, where two photons are absorbed by the material and a photon, at twice the energy of the incoming photon, is emitted. This process, due to its sensitivity to the symmetry of the system is often used as a probe for studying **surfaces and interfaces**.

However, the presence of a static electric field inside a material also enables second harmonic generation, through a third order process, named EFISH (Electric Field Induced Second Harmonic). The second harmonic spectroscopy in structures where electric fields are present becomes tricky, since we have to separate the contributions of the second order from those of the EFISH.

In this thesis, we will develop theoretical approaches to study this type of non-linear properties in the context of Time-dependent Density Functional Theory (TDDFT) and associated numerical tools for the calculation of second harmonic in the presence of a static electric field (EFISH). We will focus on the case of surfaces, interfaces to study the appearance of electric fields near the surfaces of semiconductors, as well as on non-linear properties of carbon nanotubes.