Ab initio local field effects for surface second harmonic generation

Valérie Véniard¹, Nicolas Tancogne-Dejean¹, Christine Giorgetti¹

¹) Ecole Polytechnique, CNRS, CEA-DSM-IRAMIS, Université Paris-Saclay

Corresponding author: Valerie Veniard (valerie.veniard@polytechnique.fr)

A comprehensive understanding of the nonlinear optical properties of solids is crucial to improve the design and the analysis of new optical devices and provides an opportunity to search for new materials. Among these processes, Second-Harmonic Generation (SHG) is probably one of the most studied and has become, through the years a very powerful non-invasive technique to characterize materials, because of its particular sensitivity to the symmetry of a system. In materials where inversion symmetry is present, optical Second-Harmonic Generation is forbidden within the dipole approximation. But at a surface or an interface between two such materials, the inversion symmetry is broken and SHG is allowed.

Crystal local fields are generated by the induced microscopic response of the system to an external perturbation. As a consequence their effects will be particularly important close to discontinuities as interfaces or surfaces. Local fields are important for a good description of optical properties of materials, but their effects on surface SHG have never been studied.

We present here a new ab initio formalism that allows us to calculate the frequency-dependent surface second-order susceptibility within TDDFT, where the local field effects are fully included and we have applied this formalism to Silicon surfaces.