*Institute of Nanotechnology (INT)
#Institute of Theoretical Solid State Physics (TFP)
Karlsruhe Institute of Technology D-76131 Karlsruhe

Multi-scale modeling of nonlinear optical signals from molecular materials in photonic devices

B. Zerulla*, M. Krstić#, A. L. Díaz#, C. Holzer#, D. Beutel#, I. Fernandez-Corbaton*, and C. Rockstuhl*,#

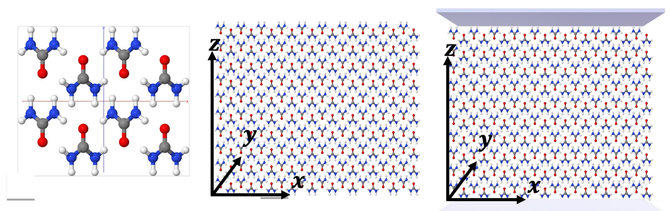
Summary

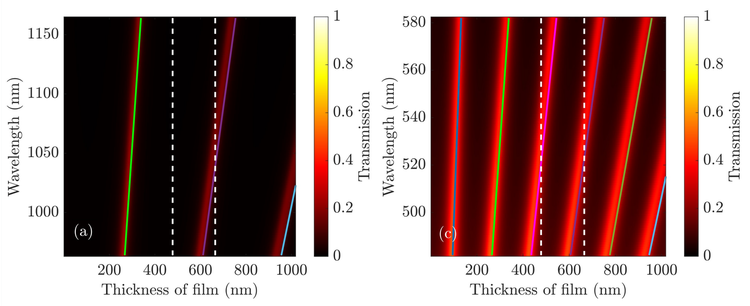
- Nonlinear optics is important for many recent photonic technologies
- We present a multi-scale approach that covers all the length scales from a single molecule to a molecular film
- It start with the *ab initio* quantum chemical computation of molecular first hyperpolarizabilities using TD-DFT in TURBOMOLE
- Uses a novel theory for multiscale simulations of nonlinear light-matter interactions based on hyper-T-matrices
- The hyper-T-matrices are obtained from the first hyperpolarizabilities, and then used in a multi-scattering code able to compute the nonlinear optical response of a single molecule, a molecular cluster, a monolayer of molecular material, or a film of molecular material, even when these are placed on top of substrates, or inside planar cavities

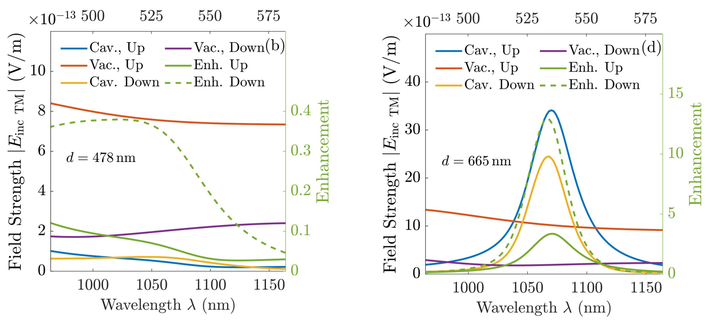
***In silico* design and optimization of devices such as optical cavities for enhancing the nonlinear signals**

Understand and quantify the second harmonic signals generated from the surface of centrosymmetric molecular crystals

Simulations of bulk SHG[1,2]

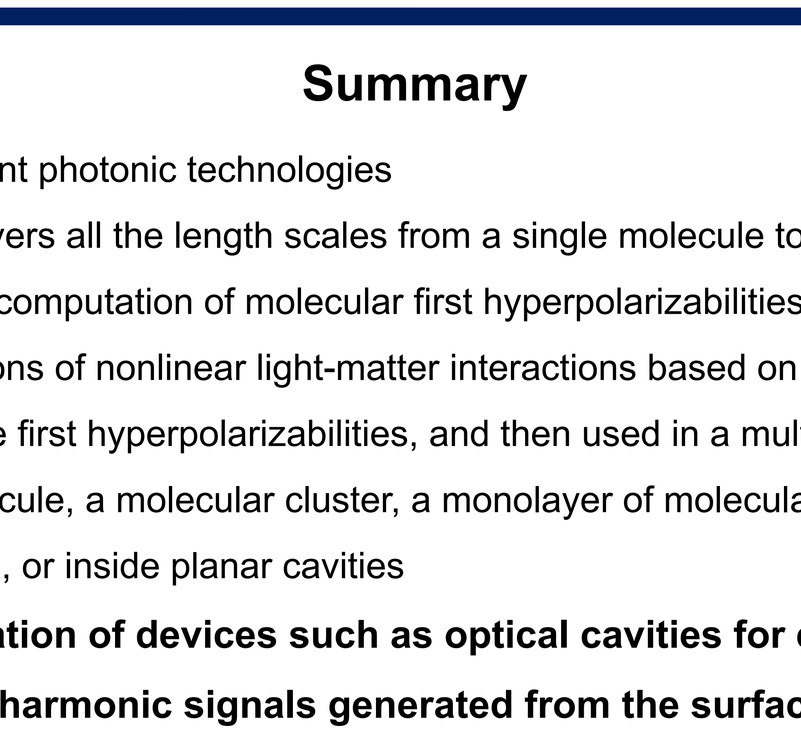


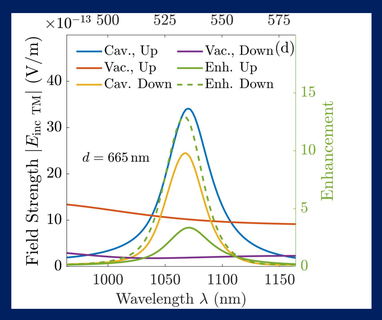


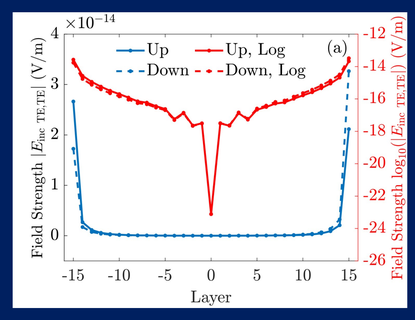


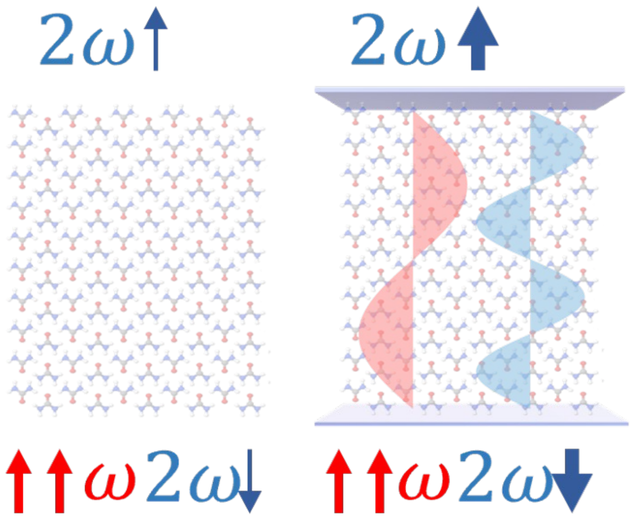
- A single-resonant cavity shows no enhancement
- A double-resonant cavity enhances the signal by a factor of 167

Surface SHG in molecular layers

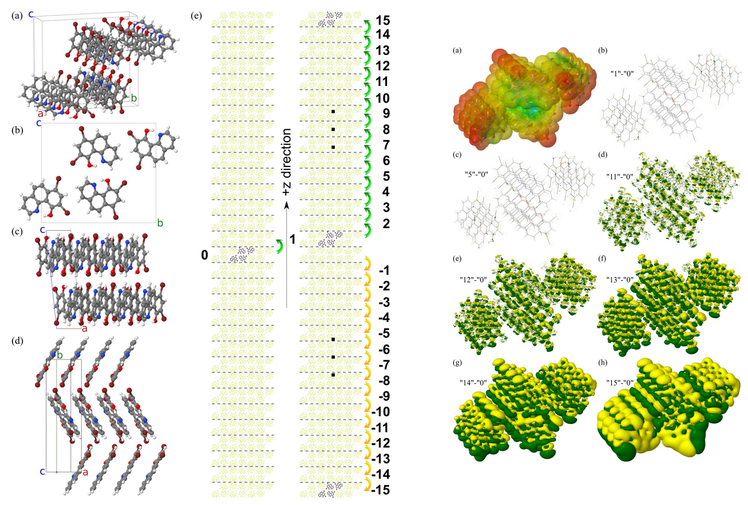








Surface SHG from layers of centrosymmetric unit cells[3]



- Symmetry-breaking deformations of the electron density revealed by embedding quantum regions within the field of partial charges

- Total internal reflection enhances the signal by factor of 1089

Outlook

- Include other nonlinear effects such as sum frequency generation, two photon absorption, third harmonic generation, and nonlinear Circular Dichroism
- Engineer the nonlinear optical response of metasurfaces and nanocavities containing molecules

1. B. Zerulla, D. Beutel, C. Holzer, I. Fernandez-Corbaton, C. Rockstuhl, M.Krstić, „A Multi-Scale Approach to Simulate the Nonlinear Optical Response of Molecular Nanomaterials", Under review, arXiv:2308.16625
2. B. Zerulla *et al.*, "A multi-scale approach for modeling the optical response of molecular materials inside cavities" *Advanced Materials Vol. 34 2200350*, **(2022)**
3. B. Zerulla, A. L. Díaz, C. Holzer, C. Rockstuhl, I. Fernandez-Corbaton, M. Krstić, „Surface Second Harmonic Generation in Centrosymmetric Molecular Crystalline Materials: How Thick is the Surface?", Submitted, arXiv:2310.20297