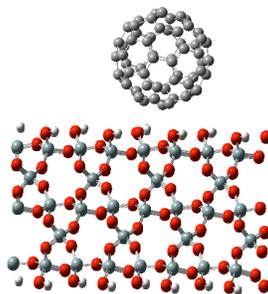# Origin of the surface-induced first hyperpolarizability in the C_{60}/SiO$_2$ system: a SCC-DFTB insight.

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In last decades, nonlinear optical (NLO) properties have been widely used to design efficient devices in different fields, like sensing, communication, or medical imaging. Nevertheless, some phenomena need further investigations to be totally understood, especially interface NLO properties. Here we investigate the second-order NLO response of buckminsterfullerene adsorbed on a silica surface. The macroscopic property of interest is the second-order NLO susceptibility, $\chi^{(2)}$, whereas at the molecular scale, the property is the first hyperpolarizability, β. Isolated C_{60} molecule is a centrosymmetric molecule and is therefore NLO inactive. However, once it is adsorbed on a silica surface, it displays a second-order NLO response. This has been observed experimentally: C_{60}/SiO$_2$ surfaces exhibit a $\chi^{(2)}$ response [1]. This result has been confirmed theoretically a few years ago, for C_{60} films as well as for single molecules adsorbed [2]. This surface-induced hyperpolarizability has first been attributed to geometric deformations or vibrational coupling at the interface.

We propose here to extend this characterization by performing quantum chemistry calculations. Owing to the size of the system, we adopt the self-consistent charge density functional tight binding (SCC-DFTB) model. This semi-empirical model permits fast calculations on periodic large systems, with a DFT-like quality. Preliminary, it was demonstrated that the SCC-DFTB method is suitable to describe the variations of first hyperpolarizabilities as a function of the structure of model compounds. Then, it is applied to the characterization of the NLO responses of the C_{60}/SiO$_2$ interface and its analysis as a function of charge transfers, energy level alignment, and polarization effects.

[1] Hoshi, H *et al.*, *Jpn. J. Appl. Phys.*, 30, L1397‑L13981, 1991.
[2] Mestechkin, M. M., *Opt. Comm.,* 273(2), 564–574, 2007.