High-throughput study of non-linear optical materials

F. Naccarato[1,2], G. Hautier[1], L. Wirtz[2] and G.-M. Rignanese[1]

[1] Université Catholique de Louvain; Institute of Condensed Matter and Nanoscience
[2] University of Luxembourg; Faculty of Science, Technology and Communication.

Nonlinear optical (NLO) processes, such as second harmonic generation (SHG), shift current, sum frequency generation, and others, play an important role in modern optics, especially in laser-related science and technology. They are at the core of a wide variety of applications ranging from optoelectronics to medicine. Among the various NLO materials, semiconductors are particularly important for second-order NLO properties. In particular, only crystals which are non-centrosymmetric can display a non-zero second-order NLO susceptibility. Furthermore, the semiconductors need to satisfy the so-called phase matching condition in order for NLO processes to occur with good efficiency. These two requirements drastically limit the choice of compounds and, despite recent progress, a systematic approach to design NLO materials is still lacking. In this work, we set up a high-throughput framework for computing the nonlinear susceptibility response using Density Functional Perturbation Theory. Within this approach, the effective SHG nonlinear coefficient is then calculated from the susceptibility tensor for a few hundred semiconductors, revealing new potential NLO materials. The data are also investigated to determine descriptors that play an important role in nonlinear phenomena.