Optical second harmonic generation (SHG) is a second-order nonlinear process by which two photons of a given frequency can combine in a nonlinear medium to generate a third photon at twice the frequency. Since the electric-dipole SHG process only occurs in non-centrosymmetric crystals, it is widely used as a sensitive probe to detect broken inversion symmetry and local polar order. Analytical modeling of the SHG response of a nonlinear optical material is essential to extract the point group symmetry as well as the absolute nonlinear susceptibilities by fitting experimental polarimetry measurements. However, complexity builds up dramatically in the analytical model when the probed crystal is of a low bulk crystal symmetry, with a surface cut in a low-symmetry orientation, exhibits absorption and dispersion, and consists of multiple interfaces.

As a result, there is a largely uneven landscape in the literature on the SHG modeling of new materials, involving numerous approximations and a wide range of (in)accuracies, leading to a rather scattered dataset of reported SHG nonlinear susceptibility. Towards streamlining the reliability and accuracy of this process, we have developed an open-source package called the Second Harmonic Analysis of Anisotropic Rotational Polarimetry (#SHAARP.si) which derives analytical solutions and performs numerical simulations of reflection SHG polarimetry from a single interface (.si) for bulk homogeneous crystals with arbitrary symmetry group, arbitrary crystal orientation, complex and anisotropic linear dielectric tensor with frequency dispersion, a general SHG tensor and arbitrary polarization of the incident and the detected SHG light.

#SHAARP.si enables accurate modeling of polarimetry measurements in reflection geometry from highly absorbing crystals or wedge-shaped transparent crystals for extracting crystal symmetry and the complete set of quantitative SHG tensor coefficients. The method is extendible to multiple interfaces.