New approach for nonlinear optical tensor determination. Application to Rare earth nickelates microcrystals

Volodymyr Multian,[1,2] Alexey Kuzmenko,[1] Dirk van der Marel,[1] and Jérémie Teyssier[1]

[1] *Department of Quantum Matter Physics, University of Geneva, 24 Quai Ernest Ansermet, CH-1211, Geneva, Switzerland*

[2] *Advanced Materials Nonlinear Optical Diagnostics lab, Institute of Physics, NAS of Ukraine, 46 Nauky pr., 03028, Kyiv, Ukraine*

We developed a novel experimental approach and data analysis technique for the reconstruction of nonlinear optical susceptibility tensors with microscopic resolution. This method is inspired by the rotational-anisotropy second harmonic generation (RA-SHG) [1] that is widely used for the readout of tensor elements of nonlinear susceptibilities of bulk materials. The standard RA-SHG mostly consists of measurements of the polarization-resolved nonlinear signal of bulk crystals and is not applicable in optical microscopes due to weak control of polarization. Our optical geometry combining a Bessel-Gaussian laser beam and a patented optical module [2] to preserve the polarization state allows us to overcome these restrictions and realize "static" RA-SHG analysis with microscopic precision and multiple polarization configurations in the same experiment. For the reconstruction of second-order nonlinear optical susceptibility tensor, we developed a fitting procedure based on evolutional algorithms that demonstrate high efficiency even for lowest-symmetry materials. This new approach is used to investigate the symmetry of the ground state in rare earth nickelates $RNiO_3$. It was predicted that this family of compounds may exhibit multiferroicity [3] and we will show that the nonlinear optical properties potentially provide experimental evidence for such behaviors.

[1] C. Yamada and T. Kimura, "Anisotropy in second-harmonic generation from reconstructed surfaces of GaAs," Phys. Rev. Lett., vol. 70, no. 15, pp. 2344-2347, Apr. 1993, doi: 10.1103/PhysRevLett.70.2344.

[2] V. Multian and J. Teyssier, "A beam splitting/mixing module for an optical system and an associated optical system," 23178209.5, Jun. 08, 2023.

[3] G. Giovannetti, S. Kumar, D. Khomskii, S. Picozzi, and J. van den Brink, "Multiferroicity in Rare-Earth Nickelates RNiO3," Phys. Rev. Lett., vol. 103, no. 15, p. 156401, Oct. 2009, doi: 10.1103/PhysRevLett.103.156401.