



Contribution ID: 258

Type: **oral**

Background-Free Electric Dipole based Chiral-HHG by Symmetry Breaking Spectroscopy: Theory and Experiment

Monday 1 July 2019 09:30 (15 minutes)

We propose and demonstrate experimentally an ultrafast chirality spectroscopy technique based on non-collinear harmonic generation, which relies only on electric-dipole interactions. The scheme is based on symmetry breaking, i.e. the chiral signal resides in harmonics that are forbidden when the medium is achiral/racemic, leading to a background-free signal. Moreover, by controlling the pump's symmetry properties the technique also yields a huge enantio-sensitive response –we experimentally measure 163% R/S selectivity.

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Session Classification: Strong field physics in topological and chiral materials