Abstract Submitted
for the MAR14 Meeting of
The American Physical Society

Non-linear optical properties derived from molecular structure via simultaneous refinement of high-resolution X-ray diffraction data and ab initio calculations JACQUELINE COLE, University of Cambridge, UK — The simultaneous refinement of experimental data and ab initio calculations is shown to afford information about the molecular origins of optical non-linearity. Specifically, non-linear optical (NLO) properties are derived from a combined experimental charge-density study, X-ray constrained wavefunction refinement, and quantum-mechanical calculations. Three case studies of well-known organic and metal-organic frequency-doubling materials highlight the power of this combined experimental and computational approach. In particular, the results show how one can derive solid-state tensorial components of molecular (hyper)polarizability directly from high-resolution X-ray structural data [1,2]. Comparing such results with those that incorporate X-ray constrained wavefunction fitting [3] demonstrate superior results. Small differences between ab initio (gas-phase) and X-ray constrained wavefunction refinement (solid-state) also reveal insights into crystal-field forces. Finally, the role of this approach in the quantum-tailored molecular design of NLO materials is forecasted.

[1] Cole et al, J. Appl. Phys. 111 (2012) 033512;
[2] Cole et al, Phys. Rev. B 004100 (2013) doi: 10.1103/PhysRevB.00.004100;
[3] Cole et al, J. Chem. Phys. 139 (2013) 064108.

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Date submitted: 15 Nov 2013 Electronic form version 1.4