Abstract Submitted
for the MAR15 Meeting of
The American Physical Society

Interplay of Atomic and Electronic Structure in Second Harmonic Generating Nonlinear Optical Materials[1] ANTONIO CAMMARATA, Czech Technical University in Prague, JAMES RONDINELLI, Northwestern University — Group theoretical methods and *ab initio* electronic structure calculations are combined to formulate a general Symmetry-Assisted Functional Optical Response (SAFOR) protocol to understand and predict the second harmonic generation (SHG) response in nonlinear optical crystals. We show that the SHG coefficients may be decomposed into atomic contributions from various inversion symmetry lifting distortions, which we parametrize as symmetry-adapted displacement patterns that transform as irreducible representations of a relevant centrosymmetric parent structure. The SAFOR protocol is then combined with an electronic descriptor for bond covalency to explain the origin of SHG in noncentrosymmetric-nonpolar $A\mathrm{TeMoO_6}$ telluromolybdate compounds. We show that the SHG response has a complex dependence on the asymmetric geometry of the polyhedral units and the orbital character at the valence band edge. The atomic scale and electronic structure understanding of the macroscopic SHG behavior obtained with these descriptions is then used to identify hypothetical $\mathrm{HgTeMoO_6}$ as a candidate telluromolybdate, which we predict should exhibit the largest SHG response in the $A\mathrm{TeMoO_6}$ family.

[1]A.C. and J.M.R. were supported by ONR and ACS-PRF under grant numbers N00014-11-1-0664 and 52138-DNI10.

Antonio Cammarata
Czech Technical University in Prague

Date submitted: 05 Nov 2014 Electronic form version 1.4