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Nonlinear Optics Simplified

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Nonlinear optical (NLO) phenomena are becoming increasingly important as diagnostics for a wide range of physical phenomena. These processes are generally treated phenomenologically in terms of components of higher-rank tensors that are allowed by symmetry. While complete, this approach fails to provide insight to the physics of NLO on the atomic scale.

We have eliminated this disadvantage and at the same time simplified the description of NLO phenomena by turned the clock back 100 years, expanding on concepts used in 1912 and 1915 by Ewald and Oseen, respectively, to describe linear optics (LO). We show that when modeled in terms of the anharmonic motion of bond charges on the atomic scale, the 4-step process of optics factors in NLO into 4 independent parts, in contrast to LO where the problem must be treated self-consistently. We are therefore led to the nonintuitive conclusion that NLO is actually simpler than LO.

Successful applications of the anisotropic bond model (ABM) and its precursors to date include second-, third-, and fourth-harmonic generation, the generation of terahertz radiation, and dipole-forbidden processes that arise in the bulk of centrosymmetric materials. Applications to glass, as a precursor to nanocrystalline Si spheres in glass, show that SHG originates from 3 distinct processes: spatial dispersion, magnetic-field effects, and retardation. The latter is a relativistic effect that is generally incorrectly described as being due to magnetic dipole/electric quadrupole interactions. SHG originating at the Si-SiO2 interface can be described completely in the ABM in terms of 3 parameters, as opposed to 11 Fourier and 14 tensor coefficients in the standard macroscopic treatment.