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Sum grequency generation spectroscopy from first principles[1]
QUAN WAN, GIULIA GALLI, Univ of Chicago — Sum frequency generation (SFG) spectroscopy is widely used to study the structural and dynamical properties of surfaces and interfaces. Within the dipole approximation, SFG signals are solely determined by the surface, and bulk contributions vanish. However, the bulk portion of a material may contribute to SFG spectra through higher multipole excitations, e.g. quadrupole, which usually are difficult to separate in the measured spectra. Here we present a first principles theoretical framework, to compute SFG spectra of molecular solids and fluids. Within the dipole approximation, we computed the dipole and polarizability using maximally localized Wannier functions (MLWF) and density functional perturbation theory [1]. We then extended our method to include quadrupole contributions, and we computed quadrupole moments and their derivatives using MLWF and a real-space correction scheme [2], and an electric enthalpy functional. We applied our approach to investigate the ice Ih surface and we present results obtained by both finite differences and ab initio molecular dynamics [3] simulations. [1] Wan *et al.* J. Chem. Theory Comput. 9, 4124 (2013) [2] Stengal *et al.* Phys. Rev. B 73, 075121 (2006) [3] Qbox Code http://eslab.ucdavis.edu/software/qbox

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