Abstract Submitted
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Sum Frequency Generation at the Al_2O_3-H_2O Interface: An Effective Bond Polarizability Model[1] MARK DELLOSTRITTO, Physics Dept., Pennsylvania State University, JAMES KUBICKI, Dept. of Geological Sciences, University of Texas at El Paso, JORGE SOFO, Physics Dept., Pennsylvania State University — Sum Frequency Generation (SFG) is a powerful tool for extracting the vibrational spectrum of an interface as it is a second-order optical process and therefore prohibited in centrosymmetric bulk media. When calculating the spectrum, it is often desirable to write the response as a sum over components of the system, such as atoms or molecules. This can pose a number challenges however, as the response depends upon the total polarizability, which is in general not an additive quantity. We employ a Thole-type model to assign polarizabilities to the bonds of a system, which allows us to treat the contribution of molecules and surface groups to the spectrum. Local field effects are then taken into account using modified Ewald sums. Following the time-dependent approach of Morita, we are able to produce the SFG spectrum at an interface from molecular dynamics simulations ranging in size and detail from small ab-initio to large classical simulations. We tested our method on ab-initio simulations of the Al_2O_3(0001)-H_2O interface as Al_2O_3 has a low dissolution rate, a well-known surface structure, and thoroughly studied surface-water interactions. We were able to successfully reproduce the experimental spectrum and decompose it in terms of molecular motions and local correlations.

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