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Effective Bond Polarizability Model for Sum Frequency Generation[1] MARK DELLOSTRITTO, JORGE SOFO, The Pennsylvania State University — Sum Frequency Generation (SFG) is a powerful, surface-specific vibrational probe ideally suited to buried interfaces, however, insight from theory is necessary to identify which surface modes give rise to the features of the vibrational spectrum. At oxide/water interfaces, strong H-bond interactions and competition between the H-bond network of water and the network of surface OH groups necessitate ab-initio MD simulations to elucidate the complex dynamics. One cannot use ab-initio methods to calculate the SFG spectrum however, due to the prohibitive cost of calculating the polarizability of large cells over long times. Thus, we develop a flexible polarizability model which takes local dipole interactions into account, rather than an additive polarizability model. We calculate bond polarizabilities and dipoles which reflect the local geometry of the H-bond network. We study the Al_2O_3 (0001)-H_2O interface, where we reproduce the experimental spectrum and show the two OH stretching peaks come from solvent and surface modes separately, not H_2O molecules with different coordination numbers as previously thought. Thus, our work emphasizes the importance of treating surface and solvent at the same level of theory for accurate spectroscopy calculations.

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