The determination of the “handedness” of chiral compounds remains a fascinating and critical challenge in which theory and computation play a vital role. In the effort to assign the absolute stereochemical configurations of chiral isolates, quantum chemical models have the potential to provide experimentalists with robust predictions of the requisite spectroscopic signatures, such as specific rotation, circular dichroism rotatory strengths, Raman scattering circular intensity differences, and more. However, such properties are among the most challenging to simulate because of their delicate dependence on a variety of intrinsic and extrinsic factors. Solvent effects, for example, not only dramatically expand the complexity of the simulation, but can sometimes even alter the sign of the chiral response. In this lecture, I will discuss recent efforts in my group toward the goal of developing reliable theoretical predictions of chiroptical properties, including the exploration of reduced-scaling methods, a variety of implicit and explicit solvation models, and even explicitly time-dependent quantum dynamics.