Molecular motion drives chemical processes. This motion is also strongly coupled to the underlying electronic rearrangements of molecules. Using renewable energy (water-splitting) catalysts and biologically relevant complexes (protonated amino acids and DNA defects) as examples, this seminar will explore the key--and often anomalous--vibrational signatures of these processes. Recent experimental developments have also motivated the need for new computational methodology that allows for reliable and intuitive explanation of anharmonic spectra. This presentation will describe a series of new methods that provide this connection and also unravel experimentally inaccessible electronic information, which can be used to explain the source of these anomalous vibrational signatures.