The function of complex molecules such as proteins depends on a delicate balance of intra- and intermolecular interactions. While the complexity of the native environment is desirable for function, isolation of biological molecules from their normal environment provides valuable simplification, enabling study of their intrinsic properties. My research program combines trapping mass spectrometry with optical spectroscopic techniques in order to characterize properties of biomolecules and their complexes in highly-controlled gas-phase micro-environments. The unique instrumentation we have developed adds powerful optical spectroscopic tools to tandem mass spectrometers, including the capability to measure fluorescence emission and Förster (or fluorescence) resonance energy transfer (FRET) of gaseous ions. I will highlight recent progress from my laboratory, discussing our insights into the intrinsic properties of a range of biological molecules and how these properties are modulated upon interaction with key binding partners. Together, this work forms a basis from which to better understand how non-covalent interactions, including those with the solvent, affect the properties of molecules ranging in size from small organic dyes to proteins.