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Enantiosensitive Photoelectron Spectroscopy of Chiral Molecules in Solution



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Life as we know it is crucially dependent on water. Biochemistry occurs nearly exclusively in aqueous environments, either under bulk solvation conditions or at aqueous interfaces. Beyond simply acting as the ubiquitous medium within which important biochemical reactions occur, water also plays a critical active role in determining the functionality of biomolecules such as amino acids, proteins, and DNA via structure stabilization and mediation of intra- and intermolecular interactions. These building blocks of life are predominantly chiral, meaning that they have a defined handedness and are non-superposable with their mirror images. Although generally chemically and physically indistinguishable, the mirror-image enantiomers of chiral molecules interact differently with other chiral entities. As a result, the activity of these enantiomers can differ dramatically under biological (chiral) conditions. Intermolecular interactions between chiral solutes and neighboring water molecules may also induce chirality in the solute's hydration shell, with profound implications for enantiomer-dependent chemistry in solution. A clearer understanding of molecular chirality in aqueous solution is thus directly relevant for chemical and life sciences.

I propose to utilize a novel and powerful technique to explore chirality in aqueous solutions of amino acids: enantiosensitive liquid-jet photoelectron spectroscopy (LJ-PES). Standard LJ-PES enables the direct study of occupied electronic states for volatile solvents and solutions with site- and chemical-state specificity. I will extend this technique by leveraging two recently-identified phenomena that highlight the photoelectron's capacity to directly access molecular chirality: photoelectron circular dichroism (PECD) and chirality induced spin selectivity (CISS). Briefly, the former effect describes the asymmetry in the angular distribution and the latter in the spin polarization of photoelectrons generated upon photoionization of chiral systems under specific experimental conditions. The use of angle-resolved PES (as required for PECD) to study liquids is uncommon, and its application to aqueous-phase chiral molecules remains at the cutting edge of molecular physics. Spin-resolved PES (the most direct means to characterize CISS) has not yet been applied to liquids in any capacity. However, the recent development of state-of-the-art LJ-PES instrumentation at the Fritz Haber Institute in Berlin has now brought the application of these techniques to chiral solutions within reach. The goal of this project is to develop and apply enantiosensitive LJ-PES to target key fundamental and technologically relevant aqueous-phase processes including induced chirality in molecular solvation shells, enantiomer-dependent photolysis and reaction kinetics, and chiral intermolecular recognition.

