Faraday Discussions

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Probing ultrafast dynamics of chiral molecules using time-resolved photoelectron circular dichroism

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Measuring the ultrafast dynamics of chiral molecules in the gas phase has been a long standing and challenging quest of molecular physics. The main limitation to reach that goal has been the lack of highly sensitive chiroptical measurement. By enabling chiral discrimination with up to several 10% of sensitivity, photoelectron circular dichroism (PECD) offers a solution to this issue. However, tracking ultrafast processes requires measuring PECD with ultrashort light pulses. Here we compare the PECD obtained with different light sources, from the extreme ultraviolet to the mid-infrared range, leading to different ionization regimes: single-photon, resonance-enhanced multiphoton, above-threshold and tunnel ionization. We use single and multiphoton ionization to probe the ultrafast relaxation of fenchone molecules photoexcited in their first Rydberg states. We show that time-resolved PECD enables revealing dynamics much faster than the population decay of the Rydberg states, demonstrating the high sensitivity of this technique to vibronic relaxation.

1 Introduction

Chiral molecules are non-superimposable to their mirror images. The two images, called enantiomers, are defined by a left or right handedness in analogy to human hands. Enantiomers have essentially the same physical and chemical properties and can only be distinguished *via* their interaction with a chiral object, such as circularly polarized light or another chiral molecule.

Terrestrial life is homochiral. All the amino acids and sugars found in living terrestrial organisms have the same handedness (L and D respectively). A

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