Abstract

The following work describes studies of proton or hydrogen transfer, one of the most basic molecular processes. Employing suitable model molecules in the form of porphycene and its derivatives as well as advanced ultrafast optical experimental methods, insight into hydrogen dynamics is offered with femtosecond temporal resolution in both ground and excited state without the requirement for spectral changes between the transfer process substrates and products, a combination of features unavailable in other techniques.

In the course of the studies, a custom experimental setup was built with improved sensitivity and flexibility over the pioneer of the technique. The construction and characterization of the apparatus as well as the workflow of obtaining hydrogen transfer rates are described. The setup was employed to study a large number of porphycene derivatives allowing, together with other experimental and computational methods, the elucidation of hydrogen transfer rate-affecting factors. Next, hydrogen tunnelling as a transfer mechanism in porphycene was investigated, with results revealing multiple tunnelling modes, including vibrationally-enhanced tunnelling significant even at room temperature. Vibrational degrees of freedom were also the focus of coherent control experiments aimed at influencing the outcome of various molecular processes in porphycenes using shaped laser pulses. Experimental realization of several useful pulse shapes is presented, as is their application to control of excited state populations and attempts to influence hydrogen transfer rates.