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Molecular Structure and Dynamics Probed by Rydberg Electrons

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— Probing molecular structure as a chemical reaction unfolds has been a long standing goal in chemical physics. Most spectroscopic and diffraction techniques work well when the molecule is cold and thus vibrational motion is minimized. Yet, in order to initiate a chemical reaction, a large amount of energy has to be pumped into the molecule. Therefore, most well-established techniques are generally inapplicable to studying ultrafast molecular transformations except for highly favorable cases. In our research we demonstrated that Rydberg electrons are very sensitive to the molecular structure. Photoionization of the molecule out of Rydberg states reveals a purely electronic spectrum not encumbered by vibrational motion. Thus, the technique is largely insensitive to the vibrations of the molecule. As an example of using Rydberg electrons as a probe for molecular structure we report on the isomerizations of cyclohexadiene and quadricyclane on an ultrafast time scale.

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