MONITORING VALENCE-ELECTRON DYNAMICS IN MOLECULES WITH ULTRAFAST X-RAY DIFFRACTION

HAIWANG YONG, STEFANO M. CAVALETTO, *Department of Chemistry, UC Irvine, IRVINE, CA, USA*;
SHAUL MUKAMEL, *Department of Chemistry, University of California, Irvine, Irvine, CA, USA*.

Ultrafast x-ray diffraction has been used to directly observe excited state electron density distributions in molecule upon photoexcitation (1). Theoretical studies have shown that its signal contains mixed elastic-inelastic coherence term originating from electronic coherence (2,3). In this study, we present a simulation study of valence-electron dynamics of oxazole using time-resolved off-resonant x-ray diffraction (4). A valence-state electronic wavepacket is prepared with an attosecond soft x-ray pulse through a stimulated resonant x-ray Raman process (5), and then probed with off-resonant single-molecule x-ray diffraction. We find that the time dependent diffraction signal originates solely from the electronic coherences and can be detected by existing experimental techniques. The present study thus provides a practical way of imaging electron dynamics in free molecules. In addition, the created electronic coherences and subsequent electron dynamics can be manipulated by resonant x-ray Raman excitations tuned to different core-excited states.

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