CFEL SCIENCE

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At Zoom virtual meeting: https://desy.zoom.us/j/83631120632

Meeting ID: 836 3112 0632 Password: 235618

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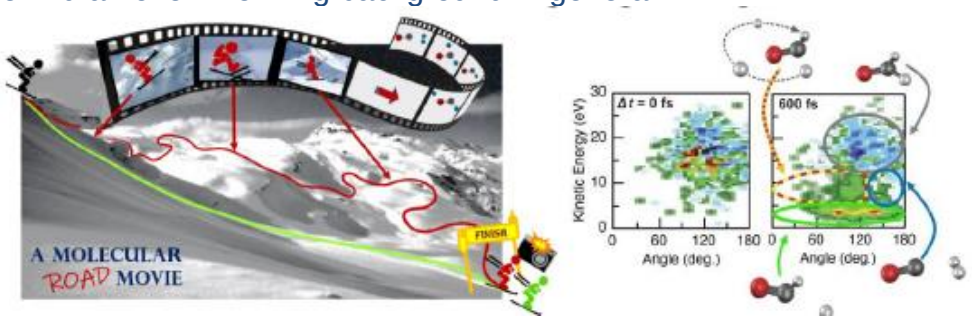
"Capturing the Statistical Roaming Signal: A Molecular Road Movie"

Synopsis Roaming molecular fragments are directly observed in the prototypical roaming reaction of the formaldehyde molecule. Despite their statistical nature, roaming fragments are well discriminated from the radical- and molecular dissociation channels, using Coulomb explosion imaging and theoretical modeling for every critical experimental step.

Since the discovery of roaming as an alternative molecular dissociation pathway in formaldehyde (H2CO) in 2004 [1], it has been indirectly observed in numerous molecules. The phenomenon describes a frustrated dissociation with fragments roaming at relatively large interatomic distances rather than following conventional transition-state dissociation; incipient radicals from the parent molecule self-react to form molecular products. Roaming has been in general identified using high-resolution spectroscopy through static product channel – resolved measurements, but not in real-time observations of the roaming fragment itself (see left schematic figure). Using time-resolved Coulomb explosion imaging (CEI), a technique sensitive to single molecules, we directly imaged individual "roamers" on ultrafast time scales in the prototypical formaldehyde dissociation reaction [2]. Using high-level first-principles simulations of all critical experimental steps, distinctive roaming signatures were identified. These were rendered observable by extracting rare stochastic events out of an overwhelming background using the highly sensitive CEI method. Also, the identification of transient features allowed to generalize the definition of roaming (see right figure). We further have started to investigate the wavelength dependence of the overall dis-sociation dynamics [3].
Here presented technique is not limited to roaming but applicable to the observation of stochastic events hidden behind an overwhelming background in general.



Left: Schematic representation of direct dissociation (green) vs. roaming (red) and the advancement of the current time resolved imaging method, compared to previous measurements once dissociation is completed. **Right**: Experimental data of total kinetic energy release vs. angle between both deuter-on momentum vectors for time zero (left) and a pump probe time delay of 600 fs (right). Areas corresponding to different dissociation pathways are indicated as: equilibrium (grey), roaming (yellow-orange dashed), molecular dissociation (blue) and radical dissociation (green).

References

[1] D. Townsend et al., *Science* **306**, 1158 (2004)
[2] T. Endo, et al., *Science*, **370**, 1072 (2020), T. Endo, et al., *Physics Today*, **07/2021**, 62 (2021)
[3] T. Endo, et al., *PCCP*, DOI: 10.1039/D1CP04264E (2022)

Host: Francesca Calegari / CFEL Molecular and Ultrafast Science Seminar