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Native frames: Separating sequential from concerted three-body fragmentation by coincidence three-dimensional momentum imaging[1]

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Advances in imaging techniques have led to better understanding of molecular fragmentation induced by photons or collisions. Experimental distinction between concerted and sequential (sometimes called stepwise) fragmentation mechanisms in polyatomic molecules is a long-standing goal. Key to its achievement is the coincidence detection of all fragments.

Using laser driven fragmentation of OCS and employing coincidence three-dimensional momentum imaging, we have recently demonstrated a novel method that enables the clear separation of sequential and concerted breakup [Phys. Rev. Lett. **120**, 103001 (2018)]. The separation is accomplished by analyzing the three-body fragmentation in the native frame associated with each step and using the rotation of the intermediate molecular fragment, before it undergoes unimolecular dissociation, as the signature of the sequential process.

Although we have demonstrated the methods benefits using OCS fragmentation by intense laser pulses, it is not limited to a specific molecule or to the interaction causing fragmentation. The method can in fact be applied to a wide variety of systems and they need not be charged that undergo three-body breakup. A sample of our recent molecular-fragmentation studies employing the Native frames analysis will be presented.

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