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Approaches for constraining uncertainty and degeneracy in geometry reconstruction of molecules from Coulomb Explosion Imaging data

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Synopsis Starting with simulated OCS molecular data we use two methods to propose a framework for reconstructing geometries, using Coulomb explosion imaging (CEI), for use in the production of molecular movies. The first method is a lookup table, which helps us investigate the existence and nature of multiple solutions to the geometry reconstruction problem and define regions of degeneracy in molecular structure. The second is a new method using nonlinear constrained optimization. We also use this method to investigate the uncertainty in geometry reconstructions as a function of measurement error.

Coulomb explosion imaging (CEI) is a technique for studying the structure and ultrafast dynamics of small molecules in the gas phase. It is possible to reconstruct the molecular structure with knowledge of the momentum vectors of every ionic fragment. We perform geometry reconstruction for simulated carbonyl sulfide (OCS) in the $OCS \rightarrow O^{2+} + C^{2+} + S^{2+}$ concerted fragmentation channel. In all cases we use a simple point like ion approach calculating final momentum vectors, for initial geometry by integrating the Coulomb repulsion to infinity. We then test the usefulness of two approaches, a look up table and a nonlinear constrained optimization approach, in terms of their speed and the uncertainty of their results.

A look up table [1] is a very simple approach for reconstructing geometries. Simulating Coulomb explosions is computationally fast, so we can simulate the explosion of many geometries and create a large mapping of geometries to asymptotic momentum vectors. Then reconstructing a geometry from measured momentum vectors is simply a matter of looking up the geometry that produces the most similar momentum vector arrangement. We test its feasibility for triatomic and larger molecules.

The nonlinear constrained optimization approach is faster and more sophisticated. To set up the statistical model, we use the interior-point method to find the geometries whose post-explosion momentum vectors most precisely match the measurements within desired constraints. This method also allows us to investigate degeneracy geometry and quantify the uncertainty on the geometry reconstructions based on the uncertainty in the momentum vectors, resulting from experimental parameters. We will therefore be able to present a two-step approach in which the look up table reveals degeneracy regions the r_{CO}, r_{CS}, θ_{OCS} space for OCS and uncertainty is dealt with by convex hulls, which are three dimensional regions representing probable values for r_{CO}, r_{CS}, θ_{OCS}, calculated using the nonlinear constrained optimization method.

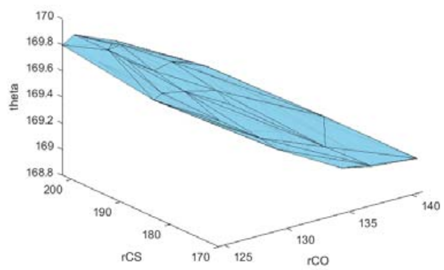

Figure1. 3D convex hull for the reconstruction of OCS (2,2,2) geometries from momentum vectors with 2% uncertainty, from the starting geometry (r_{CO}, r_{CS}, θ_{OCS}) = (130pm,190pm, 169°)

References

[1] Kunitski M 2015 *et al Science* **348** 551

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