

Ultrafast Molecular Imaging Using 4-Fold Covariance: Coincidence Insight with Covariance Speed

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Abstract: We develop mathematical tools to compute higher order covariances in charged particle detection, and demonstrate fourfold covariance measurements for molecular imaging with intense ultrafast laser pulses. © 2022 The Author(s)

1. Introduction

Momentum resolved coincidence measurements have traditionally served as the gold standard for measuring dissociative ionization of molecules. Covariance measurements can provide similar physical insights and have enabled much higher data taking rates ($\sim 100\times$) [1], allowing for studies as a function of laser intensity, pulse duration, pump-probe delay etc. While powerful, covariance was only demonstrated for up to three particles, limiting its use in Coulomb explosion imaging (CEI) of larger molecules [2]. Here we develop some mathematical tools to compute higher order covariances, and demonstrate four fold covariance for CEI of deuterated formaldehyde (CD_2O).

2. Results

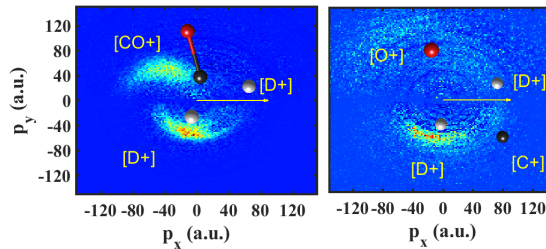


Fig. 1. Newton plots for 3-body (CO^+ , D^+ , D^+) and 4-body (O^+ , C^+ , D^+ , D^+) dissociation channels of CD_2O . The momentum distribution of all other fragment ions are displayed in the plane with one of the D^+ 's momentum fixed along the p_x direction. Both channels show similar (D^+ , D^+) angular distributions. Such an angular distribution suggests that they are ejected in a concerted manner, as discussed in detail in coincidence spectroscopy [3]. In both cases, cartoons of fragments are displayed for visual illustration.

For 3 body dissociation, Newton plots have been used in a number of molecular frame studies [3, 5]. Fig. 1 shows Newton plots for three and four body dissociation channels of CD_2O . In Fig. 1, one of the D^+ momentum is fixed as the reference for the other fragments. The left panel of Fig. 1 depicts the Newton plot for the $(\text{CO}^+, \text{D}^+, \text{D}^+)$ channel. The shape of the distribution is quite similar to that of the coincidence 3-body dissociation of $\text{C}_2\text{H}_2\text{Cl}_2$ [3]. The right panel shows the same plot for the 4-body channel $(\text{O}^+, \text{C}^+, \text{D}^+, \text{D}^+)$. Although the statistics are limited, the momentum of the second D^+ can still be seen, which suggests it is ejected in a process similar to the 3-body dissociation.

While it is tempting to consider fourfold covariance in terms of an extension of expressions for lower orders:

$$\begin{aligned} \text{Cov}(A, B, C) &= \langle (A - \langle A \rangle)(B - \langle B \rangle)(C - \langle C \rangle) \rangle \\ \text{Cov}(A, B, C, D) &= \langle (A - \langle A \rangle)(B - \langle B \rangle)(C - \langle C \rangle)(D - \langle D \rangle) \rangle, \end{aligned} \quad (1)$$

it can be shown that this does not produce physical results without corrections. Fig. 2 illustrates the direct application of this expression along with the corrected version. Fig. 2 shows a test of momentum conservation with covariance analysis for partial $(\text{O}^+, \text{C}^+, \text{D}^+)$ and full 4-body dissociation channels $(\text{O}^+, \text{C}^+, \text{D}^+, \text{D}^+)$. The x component of the momentum (p_x) for the fragments are summed. For the partial channel, represented by the green curve, momentum conservation fails due to the lack of momentum information of the missing D^+ (or D) fragment. For the full (fourfold) channel, the uncorrected formula given above yields the dark red curve, which shows a peak around zero momentum. However, in addition to the peak at zero, there is a broad background distribution over a wide range of momentum. This problem is completely solved by utilizing the appropriate correction. The result of the corrected analysis yields the red curve in the figure, which simply shows a clean peak around zero momentum, emphasizing the importance of the correction.

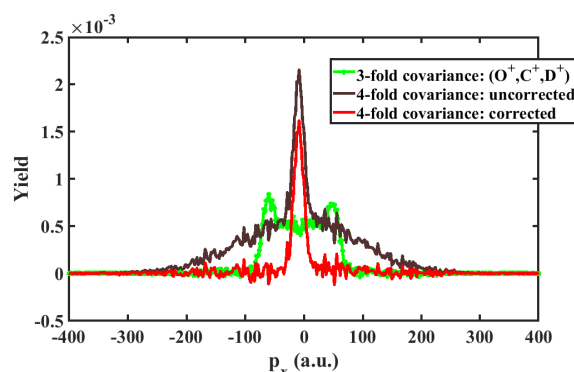


Fig. 2. Histogram of sum of momenta in x direction for different dissociation channels. The green curve shows that momentum is not conserved due to the missing D^+ (or D) fragment. The light red curve represents the 4-fold covariance histogram using the correct formula. The curve shows a single peak around zero momentum with limited fluctuations, indicating that momentum is conserved in covariance, as it is in coincidence [4]. In contrast, the dark red curve, using the normal covariance formula, shows not only a sharp peak at around zero momentum but also a large background. Such a difference illustrates the importance of the correction to the covariance calculation.

References

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