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Adaptive femtosecond control using feedback from three-dimensional momentum images[1]

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Shaping ultrafast laser pulses using adaptive feedback is a proven technique for manipulating dynamics in molecular systems with no readily apparent control mechanism. Commonly employed feedback signals include fluorescence or ion yield, which may not uniquely identify the final state. Raw velocity map images, which contain a two-dimensional representation of the full three-dimensional photofragment momentum vector, are a more specific feedback source. The raw images, however, are limited by an azimuthal ambiguity which is usually removed in offline processing. By implementing a rapid inversion procedure based upon the onion-peeling technique, we are able to incorporate three-dimensional momentum information directly into the adaptive control loop. This method enables more targeted control experiments. Two examples are used to demonstrate the utility of this feedback. First, double ionization of CO produces C^+ and O^+ fragments ejected both perpendicular and parallel to the laser polarization with kinetic energy release of $\sim$6 eV. Both suppression and enhancement of the perpendicular transitions relative to the parallel transitions are demonstrated. Second, double ionization of acetylene can lead to both $HCCH^{2+}$ and $HHCC^{2+}$ isomers. We select between these outcomes using the angular information contained in the CH^+ and CH_2^+ images.

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