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Molecular Recollision Interferometry in High Harmonic Generation XIBIN ZHOU, ROBYNNE LOCK, NICK WAGNER, WEN LI, MARGARET MURNANE, HENRY KAPTEYN, JILA and Department of Physics, University of Colorado and NIST, Boulder, CO 80309, USA — There has been considerable recent interest in using high-order harmonic generation to observe molecular structure and dynamics. Besides measuring the intensity of high-order harmonics, we use extreme-ultraviolet interferometry to measure the phase of high-order harmonic generation from transiently aligned CO₂ molecules. We unambiguously observe a reversal in phase of the high order harmonic emission for higher harmonic orders with a sufficient degree of alignment. This results from molecular-scale quantum interferences between the molecular electronic wave function and the recolliding electron as it recombines with the molecule, and is consistent with a two-center model. Furthermore, using the combined harmonic intensity and phase information, we extract accurate information on the dispersion relation of the returning electron wave packet as a function of harmonic order. This analysis shows evidence of the effect of the molecular potential on the recolliding electron wave. Our measurements are critical for developing new approaches for molecular imaging.

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