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High-harmonic Spectroscopy of Two-center Interference in Aligned OCS and CO2¹ TIMOTHY GORMAN, TIMOTHY SCARBOROUGH, Department of Physics, The Ohio State University, PETER SANDOR, SANJAY KHATRI, Department of Physics, University of Virginia, FRANCOIS MAUGER, PAUL ABANADOR, Department of Physics and Astronomy, Louisiana State University, ROBERT JONES, Department of Physics, University of Virginia, METTE GAARDE, KENNETH SCHAFER, Department of Physics and Astronomy, Louisiana State University, PIERRE AGOSTINI, LOUIS DIMAURO, Department of Physics, The Ohio State University — We present high harmonic spectroscopic measurements on the spectral intensity and phase of two-center interferences resulting from laser-driven electron-core recollisions in OCS and CO2. Using a long-wavelength 1.3 um driving laser for high harmonic generation combined with impulsive molecular alignment we are able to reveal a change in both the sign and magnitude of the spectral phase jump between the two molecules. Additionally, we are able to control this interference by rotating the molecular axis relative to the laser polarization, demonstrating clear differences between the angular dependencies of the two molecules. Our study offers a precise comparison between molecular-frame interferences of polar and nonpolar molecules, potentially important for future studies of attosecond charge migration and coherent control of molecular processes.

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