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Polarization Resolved Characterization of High Harmonic Emission from Aligned Molecules XIBIN ZHOU, ROBYNNE LOCK, HENRY KAPTEYN, MARGARET MURNANE, JILA and Department of Physics, University of Colorado and NIST — High-order harmonic emission from molecules that have been impulsively aligned can be used to obtain information on molecular structure and ultrafast dynamics. Complementary to the intensity measurement, the polarization properties of this emission can offer considerably more insight and can provide for critical tests of the theory of molecular high harmonic generation (HHG). In this work, we present the results of two experiments. First, we perform accurate polarimetry measurements of the HHG emission of aligned N_2 and CO_2 [1]. We find that in N_2 , the HHG emission can be strongly elliptically polarized even when the laser used to drive the process is linearly polarized. In contrast, the harmonic emission from aligned CO_2 molecules shows a polarization rotation but no significant ellipticity. In the second experiment, we vary the ellipticity of the driving laser used to generate the high harmonic emission. We find that the HHG intensity from aligned molecules does not peak for linear polarization of the driving laser (as in the atomic case), but at a small positive or negative ellipticity. The sign and degree of this ellipticity depend on the molecular orientation. [1] X. Zhou et. al., PRL accepted (2009).

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