Polarization Characterization of High Harmonic Generation from Transiently Aligned Molecules

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Synopsis: We perform an accurate polarimetry measurement of high harmonic emission from aligned molecules. We find that harmonic emission from nitrogen molecules can be strongly elliptically polarized; even when driven by linearly polarized laser fields. We also 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 optimize with linear polarization of the driving laser (as in the atomic case), but rather peaks at a small positive or negative ellipticity. The sign and degree of this ellipticity depend on the molecular orientation.

High-order harmonic generation (HHG) results from the extreme distortion of an electron wave function in an atom or molecule in the presence of a strong laser field. In the case of HHG from molecules, the intensity and phase of the EUV light can encode structural information - particularly if the molecular sample is aligned and thus this information is not averaged-out.

In principle, the amplitude, phase and polarization of HHG emission from molecules can be measured and used to extract the electronic orbital structure from the transition dipole matrix element of the recombination step of the HHG process. However, until very recently, only the intensity of HHG from molecules could be measured. Fortunately, new approaches have made it possible to determine the phase of HHG. In this abstract, we present two experiments where the polarization state of high harmonics from an ensemble of aligned molecules is characterized.

In the first experiment, we perform the first accurate polarization measurement of the ellipticity of HHG emission from molecules [1]. We extract the phase difference between the HHG components parallel and perpendicular to the driving laser field, which depends only weakly on molecular alignment but strongly on the harmonic order. Our findings differ from previously published results [2], where only linearly-polarized harmonic emission was observed, because we obtain a stronger molecular alignment and better signal-to-noise ratio (1~2% uncertainty). This allows us to detect ellipticity in the HHG emission even when molecules are driven by linearly-polarized laser fields. These findings cannot be explained by a plane-wave SFA, and will be very useful to benchmark new and more complete theories of molecules in strong fields.

From the measured ellipticity and orientation angle of the HHG polarization ellipse, we can extract the phase difference and amplitude ratio of the orthogonal HHG polarization components. The observed nontrivial variation in relative phase indicates a breakdown of the plane wave approximation. In the second experiment, we varied the ellipticity of the driving laser used to generate the high harmonic emission. We find that the HHG intensity generated from an ensemble of 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 depends on the molecular orientation. The most probable explanation for this behavior is the manipulation of the returning electron wave packet by the elliptical polarization of the laser.

In summary, we have uncovered unexpected polarization properties of high harmonic emission from N_2 and CO_2 molecules that have broad implications for the theory of molecules in strong fields. Furthermore, we show that HHG from aligned molecules driven by an elliptically-polarized laser pulse can also yield new information information. More-sophisticated models of the laser molecule interaction will be required to fully understand these experimental data.

References

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