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Revealing molecular structure and dynamics through high harmonic generation driven by mid-IR fields[1]

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High harmonic generation (HHG) from molecules has recently been shown to be a promising tool for measuring instantaneous molecular structure, sub-femtosecond domain structural rearrangements in molecules and even hole dynamics initiated by laser field ionisation. To fully exploit this promise it is essential that we can; (1) systematically decouple structural and dynamic effects so that both may simultaneously be determined in the measurement, (2) can extend the method of molecular HHG imaging to a wide range of molecules. Here we demonstrate important steps towards both these objectives. Up until now HHG imaging measurements have been restricted to drive laser wavelengths close to 800nm, due to the availability of CPA titanium sapphire lasers, which dictates the use of relatively high intensities (> 2.5 x 10^{14} Wcm^{-2}) if a harmonic spectrum spanning to $\sim$70 eV is to be observed which is required for extracting structural data from most small molecules. By using a mid-IR laser (at 1300 nm) we show that with an intensity ~ 1 x 10^{14} W cm^{-2} we can observe a wide molecular harmonic spectrum spanning to $\sim$ 70 eV even in molecules where ionization saturation would clamp the cut-off to much lower energies if an 800nm field were used. Thus we have been able to observe evidence for two-centre interference in two new molecules, N_2O and C_2H_2 for the first time. Moreover we can use the ability to observe a broad harmonic spectrum over a large range of intensities to reveal the subtle interplay between structural and dynamic effects in CO_2 and so provide a new window into hole dynamics.

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