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Synopsis We present experimental results of linear-to-bent transition of field-dressed molecules, mediated by Renner-Teller effect. Using the state-of-the-art laser-induced electron diffraction (LIED) technique, we image a bent and symmetrically stretched carbon disulfide (CS_2) molecule populating an excited electronic state under the influence of strong laser field. Our findings are well-supported by *ab initio* quantum mechanical calculations.

High-resolution ultrafast dynamics of structural changes are essential in the real-time observation of chemical reactions. Here, we observe ultrafast structural changes in CS_2 with a single-pulse excitation and imaging under the influence of strong laser field [1].

The experiment was performed using the LIED technique [2, 3] with the help of a passively CEP-stable mid-IR OPCPA laser system (3.2µm, 110fs, 160 kHz) [4] coupled with a reaction microscope [5], the latter of which is capable of measuring the full three-dimensional momenta of all charged particle species.

LIED probes an object's structure using its own electrons that are elastically re-scattered during strong-field induced re-collisions, as explained by the semi-classical re-collision model [6]. The excursion time of the rescattering electron and its high momentum transfer (~ 9.5 A^{-1}) provide a sub-femtosecond temporal resolution and picometre spatial resolution, respectively.

Our experimental findings show that the vibronic excitation of neutral CS_2 to a bent and symmetrically stretched structure occurs during the rising edge of our LIED laser field, as illustrated in Fig. 1. Our *ab initio* quantum dynamical calculations show that the measured bent structure in CS_2 in the presence of strong field is due to a linear-to-bent structural change mediated by the Renner-Telller effect, and is a consequence of the coupling of rovibronic states to the electronic states.

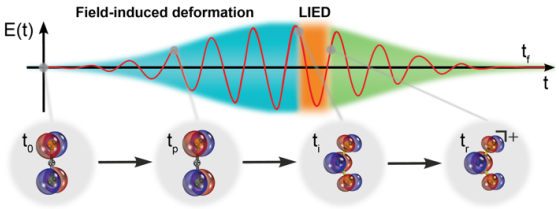

Figure 1. The CS_2 molecule is initially stretched and bent by 10° (at t_p) before leading to a bent CS_2 structure that is ionized (at t_i) and imaged by high-energy re-scattering electrons (at t_r).

In future work, LIED can be extended to two-pulse pump-probe studies to investigate the pathways of structural photo-induced chemical reactions suh as those in the photoisomerization of photoswitching molecules.

References

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