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Polyatomic molecular structure retrieval using laser-induced electron diffraction

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Synopsis Laser-induced electron diffraction is a developing dynamical imaging technique that is already able to probe molecular dynamics at few-femtosecond temporal resolutions and has the potential to reach the sub-femtosecond level. Here we provide the recipe for the extension of the technique to polyatomic molecules and we demonstrate the method by extracting the structure of aligned and anti-aligned acetylene (C_2H_2). We show that multiple bond lengths can be simultaneously imaged at high accuracy including elusive hydrogen containing bonds. Our results open the door to the investigation of larger complex molecules and the realization of a true molecular movie.

Conventional electron diffraction (CED) is able to determine internuclear distances at unsurpassed spatial accuracies, however, pushing the achievable temporal resolutions below a few-hundred femtoseconds has proved difficult. On the other hand, few-femtosecond structural dynamics of the O_2 molecule have already been reported using laser-induced electron diffraction (LIED) [1]. However, the real goal of LIED research is to probe polyatomic molecules that have many possible fragmentation channels. Disentangling information from these concurrent processes requires many capabilities that standard experimental apparatuses do not possess. Additionally, the low ionization potential of polyatomic molecules places strict limitations on the driving radiation. Therefore, up until now, LIED has been limited to diatomic molecules. Here, we provide the recipe to overcome these non-trivial hurdles and present the first simultaneous extraction of multiple bond lengths in aligned and anti-aligned acetylene molecules (C_2H_2). Moreover, we show how the electron recollisions induced by strong-field ionization can potentially be used to resolve sub-femtosecond dynamics.

Our experimental apparatus is composed of a combination of an intense 160 kHz, 3.1 µm source with a reaction microscope (ReMi) detection system. The mid-IR radiation generates high kinetic energy core penetrating re-scattering electrons and the ReMi is able to detected both electron and ion momentum distributions in full coincidence. Molecular structure information is hidden in the electron momentum distribution (Fig. 1) and can be retrieved following the quantitative re-scattering (QRS) theory. The obtained bond lengths agree well with the expected values for both alignments.

To our knowledge, our results show the first simultaneous extraction of multiple bond lengths from a polyatomic molecule using LIED [2]. Our unique experimental apparatus opens up the possibility of applying LIED to much larger and more complex molecules. Additionally, information about fragmentation processes is already available due to the coincidence capabilities of the ReMi detection system. Combining our methodology with the already presented few-femtosecond capabilities of LIED truly represents a realistic path to achieving the long sought after 'molecular movie'.

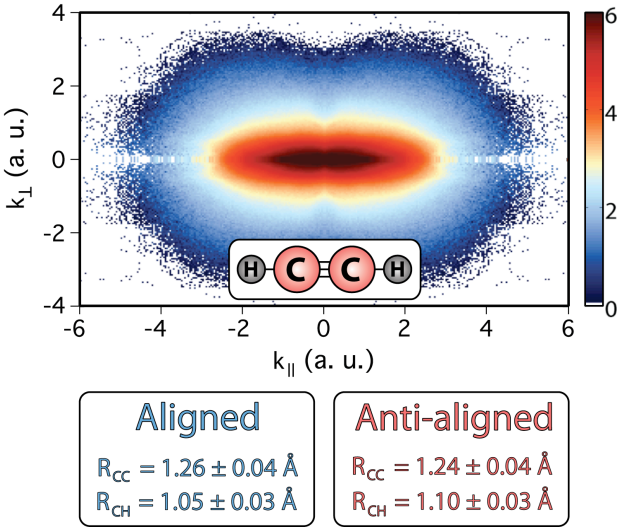

Figure 1. Electron momentum distribution (log scale) detected by the ReMi after the ionization of aligned C_2H_2 molecules by the high repetition rate 3.1 um source. Accurate C-C and C-H bond lengths can be simultaneously extracted from this diffraction pattern for both aligned and anti-aligned molecules.

References

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