

Structural retrieval of C₂H₄ molecules by laser induced rescattering photoelectron spectroscopy

Yuta Ito*, Richard Carranza[†], Misaki Okunishi*, Robert R. Lucchese[†], Kiyoshi Ueda^{*1}

* Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan

[†] Department of Chemistry, Texas A&M University, College Station, Texas 77843-3255, USA

Synopsis We measured angle-resolved rescattering photoelectron spectra of C₂H₄ molecules, from which electron-ion differential scattering cross sections were extracted. We did an ab initio calculation for elastic e⁻-C₂H₄⁺ scattering, using ePolyScat, a scattering calculation program suite. By comparing the experimental and theoretical results, bond lengths were retrieved for both C=C and C-H accurately.

A growing number of studies have been carried out to establish new experimental techniques to map a structural change of gas phase molecules in a femtosecond time scale. Rescattering electron spectroscopy in intense laser fields is one of the methods for such a purpose. After a proposal of extraction method of differential cross sections (DCSs) for electron-ion elastic scattering from the rescattering photoelectron spectra [1, 2], a few number of studies have been reported on structure retrieval of linear [3, 4] or planar [5] molecules. In this study, we measured angle-resolved rescattering electron spectra of C₂H₄ and compared the results with theoretical calculations employing ePolyScat of a scattering calculation package toward the structure retrieval.

The output of a Ti:Sapphire laser (800 nm, 100 fs, 1.5 mJ, 1 kHz) was injected to an optical parametric amplifier to convert the wavelength to 1650 nm. The laser was introduced in a high vacuum chamber through a half waveplate and focused on C₂H₄ molecules. Emitted electrons were detected with a field-free time-of-flight spectrometer, as rotating the polarization of the laser to get a two-dimensional momentum distribution of the electrons.

DCSs (σ_M) were extracted from the experimental spectra for recollision momentum (p_r) between 2.0-2.5 a.u. In Fig. 1, molecular contrast factors (MCFs) defined as $(\sigma_M - \sigma_A) / \sigma_A$ are depicted as a function of momentum transfer, where σ_A is theoretical atomic DCSs, or the incoherent sum of DCSs for all the atoms in the molecule. A clear oscillatory structure, which may reflect molecular geometry, can be seen, showing good agreement with a theoretical one. Residual sum of squares were obtained between experimental and theoretical MCFs, changing the C=C and C-H bond lengths for theoretical ones. In Fig. 2, the results show minima for both C=C and C-H, where we get the value of 1.32 Å and 1.04 Å, respectively. They are very close to ones for the equilibrium geometry (1.34 Å for C=C and 1.09 Å for C-H) within ~5 % of difference, indicating that the

structural retrieval could be successfully performed.

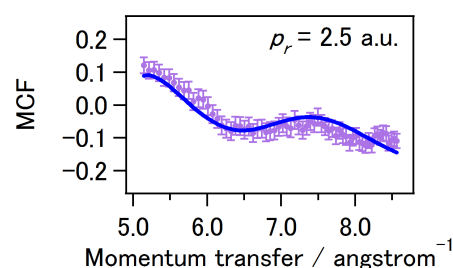


Figure 1. Experimental MCF (violet) and theoretical MCF with ePolyScat (blue).

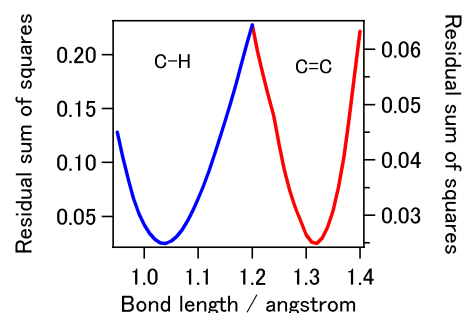


Figure 2. Residual sum of squares between experimental and theoretical MCFs as a function of C=C (red) or C-H (blue) bond lengths.

This work was supported in part by Grants-in-Aid for Scientific Research from JSPS, by the X-ray Free Electron Laser Utilization Research Project and the X-ray Free Electron Laser Priority Strategy Program of the MEXT-Japan and IMRAM research program.

References

- [1] Morishita T *et al.* 2008 *Phys. Rev. Lett.* **100** 013903
- [2] Chen Z *et al.* 2009 *Phys. Rev. A.* **79** 033409
- [3] Blaga C I *et al.* 2012 *Nature* **483** 194
- [4] Pullen M G *et al.* 2015 *Nat. Commun.* **6** 7262
- [5] Ito Y *et al.* 2016 *Struct. Dyn.* **3** 034303

¹E-mail: ueda@tagen.tohoku.ac.jp