

Laser-assisted electron scattering and diffraction for ultrafast molecular imaging

Reika Kanya¹, Yuya Morimoto, Kakuta Ishida, and Kaoru Yamanouchi²

Department of Chemistry, School of Science, the University of Tokyo, Tokyo 113-0033, Japan

Synopsis Techniques for ultrafast molecular imaging based on laser-assisted electron scattering (LAES) processes are reported. We demonstrate a method for achieving high temporal resolution in gas electron diffraction by detecting 1 keV electrons scattered by CCl₄ in a femtosecond laser field, and show that the geometrical structure of molecules at the instant of the laser pulse irradiation can be determined by this method. Furthermore, we observed multiphoton free-free transitions in LAES by Xe atoms in a femtosecond near-infrared intense laser field, and showed that angle-resolved energy spectra of the LAES signals exhibit clear plateau structures, in which ultrafast dynamics in the sub-fs time scale is encoded.

When an electron is scattered by an atom or a molecule in a laser field, the electron can change its kinetic energy by multiples of the photon energy ($\hbar\omega$). This scattering process is called laser-assisted electron scattering (LAES) or free-free transition of electrons. Recently we proposed an ultrafast electron diffraction method called laser-assisted electron diffraction (LAED) [1] as an application of the LAES process. Because the LAES process occurs only in the presence of the laser field, the temporal resolution of the LAED method can be as short as the laser pulse duration (< 10 fs). In the present study, we recorded LAED patterns appearing through the interference among LAES electrons scattered by the respective atoms within a molecule at the energy shift (ΔE) of $\pm\hbar\omega$ through the scattering of a 1 keV electron beam by CCl₄ molecules in a femtosecond intense laser field ($\lambda = 800$ nm, $\Delta t = 520$ fs, $I = 6 \times 10^{11}$ W/cm²) [2]. Through the comparison with the numerical simulations, we show that the LAED pattern carries the information of the geometrical structure of molecules during the ultrashort duration of the laser irradiation.

The angular distribution of LAES signals of $\Delta E = +\hbar\omega$ is shown in Fig. 1 as filled circles. In the angular distribution, we observed clear interference structures with a minimum around 5.5° and a maximum around 9.0°. In order to confirm the origin of the interference patterns, we conducted a numerical simulation based on the Kroll-Watson theory [3]. The simulated LAES angular distribution represented by the solid curve in Fig. 1 is in good agreement with the experimental distribution. This agreement shows that the observed interference patterns are the electron diffraction patterns of CCl₄.

The present study shows that the geometrical structure of CCl₄ can be determined from the analysis of the LAED signals. By using few-cycle laser pulses, temporal resolution of sub-10 fs will be achieved.

We also performed recently measurements of high-order multiphoton LAES processes by Xe atoms and showed that plateau structures appear in angular resolved energy spectra of scattered electrons [4], from which we can discuss ultrafast dynamics of electrons colliding with a target atom in the sub-femtosecond time scale based on a classical mechanical model.

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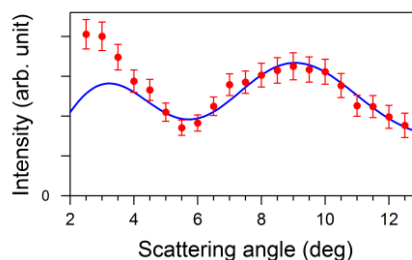


Figure 1. Angular distribution of LAES signals of $\Delta E = +\hbar\omega$. Filled circles: experimental data, Solid line: simulation.

References

- [1] R. Kanya, Y. Morimoto, K. Yamanouchi, 2010 *Phys. Rev. Lett.* **105** 123202
- [2] Y. Morimoto, R. Kanya, K. Yamanouchi, 2014 *J. Chem. Phys.* **140** 064201
- [3] N. M. Kroll, K. M. Watson, 1973 *Phys. Rev. A* **8** 804
- [4] K. Ishida, Y. Morimoto, R. Kanya, K. Yamanouchi, 2017 *Phys. Rev. A* **95** 023414

¹ E-mail: kanya@chem.s.u-tokyo.ac.jp

² E-mail: kaoru@chem.s.u-tokyo.ac.jp