Ultrafast gas electron diffraction by THz-wave assisted electron scattering: Numerical simulations of time-resolved electron diffraction patterns

Reika Kanya [1] **and Kaoru Yamanouchi** [2]

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

Synopsis A method for achieving high temporal resolution in gas electron diffraction without scanning the pump-probe delay is proposed. In the proposed method, time-dependent electron diffraction patterns can be obtained from energy-resolved angular distributions of electrons scattered by molecules in dynamical processes under the presence of a single-cycle THz wave pulse. Derived formulae of the differential cross section and numerical simulations of electron signals scattered by Cl_2 molecules show that the temporal resolutions of the proposed method can be better than 10 fs.

One of the most challenging themes in molecular science is to probe ultrafast changes in geometrical structures of isolated molecules in real time with the atomic-scale spatial resolution. Until now several pioneering attempts have been made for achieving the goal by using newly developed experimental methods such as electron diffraction with ultrashort relativistic electron pulses, X-ray diffraction with X-ray free electron lasers, laser-induced electron diffraction, and laser-assisted electron diffraction as reviewed in Ref. [1]. In the present study, we propose another electron diffraction method called THz-wave assisted electron diffraction (TAED). In TAED method, by using half-cycle THz-wave pulses as streaking electric fields affecting the electron diffraction process, a series of snapshots of electron diffraction patterns of isolated molecules can be obtained with femtosecond temporal resolutions, that is, dynamical processes of molecules can be recorded in real time without scanning the pump-probe delay time.

First, the Kroll-Watson theory [2], which has been regarded as a standard theory for LAES processes, was modified so that it can be applied even for ultrabroadband electromagnetic pulses. The obtained formula of differential cross sections for the LAES processes induced by half-cycle THz-wave pulses shows that the time when the incident electron collides with the target atom or molecule can be determined by the energy shift and the momentum transfer of the scattered electron. By using the formula, numerical simulations were performed for calculating electron scattering intensities by Cl_2-like molecules in a half cycle THz pulse whose peak electric field intensity and peak frequency are 80 kV/cm and 1.0 THz, respectively. Figure 1 shows the simulated electron scattering signals for dissociating Cl_2 molecules. By transforming the energy shift axis to the collision time axis, it was confirmed that the electron diffraction pattern varying in real time can be obtained by the TAED measurement without scanning pump-probe delay. From the obtained time-dependent diffraction pattern, the internuclear distances as a function of time were retrieved with the accuracy better than 1.5×10^{-3} Å. Assuming that the spatial variation of the electric field strength of the THz wave at the scattering point is around 3%, the temporal resolution is estimated to be better than 10 fs, showing that the geometrical structures of molecules can be determined by the TAED method with the temporal resolution of sub-10 fs and the spatial resolution of around 10^{-3} Å.

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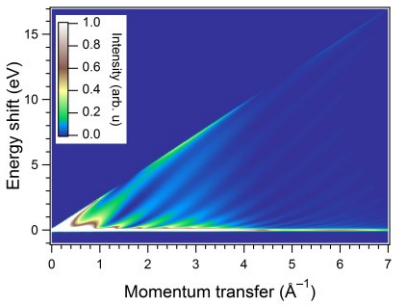

Figure 1. Signal distributions of electrons scattered by dissociating Cl_2 molecules.

References

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[1] E-mail: kanya@chem.s.u-tokyo.ac.jp

[2] E-mail: kaoru@chem.s.u-tokyo.ac.jp