Determination of collision times in laser-assisted electron scattering for ultrafast imaging of atoms and molecules

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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. In 2014, we showed that laser-assisted electron diffraction [1] is one of the promising methods to achieve the femtosecond temporal resolution in the determination on geometrical structures of molecules. Recently, we proposed another electron diffraction method called THz-wave assisted electron diffraction (TAED) [2] and showed that instantaneous geometrical structures of molecules can be determined with the femtosecond temporal resolution by estimating collision times from the energy-resolved angular distribution of electrons streaked by laser-assisted electron scattering (LAES) processes induced by single-cycle THz-wave pulses. We also introduced a scheme with which we can estimate the collision times in the LAES processes by Xe induced by multi-cycle near-infrared laser pulses with the attosecond resolution [3]. In the present study, we show through numerical simulations that the temporal variation of the internuclear distance of H_2^+ molecular ions can be determined with the attosecond temporal resolution by the LAES in a single-cycle mid-infrared laser field.

Using a formula of the differential cross section of LAES processes induced by broadband electromagnetic pulses [2], we calculate signal distributions of electrons scattered by dissociating H_2^+ in a single-cycle mid-infrared laser pulse (λ = 4 μm, Δt = 13 fs, I = 1.0×10^{11} W/cm^2). In the simulation, the H-H molecular axis is set to be parallel to the laser polarization direction, and the direction of the incident electron beam is set to be perpendicular to the molecular axis. Figure 1(a) shows the temporal shape of the laser field, and Fig. 1(b) shows the signal distributions of electrons scattered by dissociating H_2^+. From the analysis of the streaked electron diffraction patterns, the time-dependent internuclear distance, $R(t)$, of H_2^+ is retrieved. As shown in Fig. 1(c), the retrieved $R(t)$ (the gray solid line) is in good agreement with the initially given $R(t)$ (the black broken line), showing that ultrafast structural changes in geometrical structure of molecules can be probed with an attosecond temporal resolution.

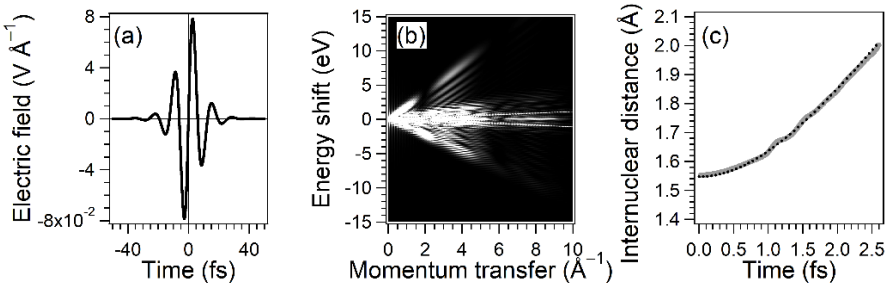

Figure 1: (a) Temporal shape of the ultrashort mid-infrared laser pulses adopted in the present simulation. (b) Signal distributions of electrons scattered by dissociating H_2^+. (c) Time-dependent internuclear distance, $R(t)$. Black broken line: initially given $R(t)$, Gray solid line: $R(t)$ retrieved from the analysis of the intensity distributions in (b).

[1] Morimoto Y, Kanya R and Yamanouchi K 2014 *J. Chem. Phys.* **140** 064201
[2] Kanya R and Yamanouchi K 2017 *Phys. Rev. A* **95** 033416
[3] Ishida K, Morimoto Y, Kanya R and Yamanouchi K 2017 *Phys. Rev. A* **95** 023414