GEC15-2015-000071

Abstract for an Invited Paper
for the GEC15 Meeting of
the American Physical Society

Progress in (e, 2e) electron momentum spectroscopy: from the static to the time-resolved regime

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Electron momentum spectroscopy (EMS) is a kinematically-complete electron-impact ionization experiment performed under the high-energy Bethe ridge conditions, where the collision kinematics can be described by electron Compton scattering that most nearly corresponds to the collision of two free electrons with the residual ion acting as a spectator. The remarkable feature of this technique is its ability to measure momentum distributions of each electron bound in matter or to look at molecular orbitals in momentum space. We have been exploring atomic and molecular science using EMS, such as 3D orbital imaging for a stable gaseous molecule [Takahashi et al., PRL 2005], observation of the giant resonance phenomenon in the 2nd order projectile-target interactions [Takahashi et al., PRL 2007], and determination of spatial orientation of the constituent atomic orbitals in molecular orbitals [Watanabe et al., PRL 2012]. Recently, we have started to direct our efforts also towards expanding frontiers of EMS, through development of time-resolved EMS (TR-EMS) that employs ultrashort laser (120 fs) and electron (1 ps) pulses in a pump-probe scheme [Yamazaki et al., RSI 2013]. In spite of the low data statistics as well as of the limited time-resolution due to velocity mismatch, our experimental results on the deuterated acetone molecule in its second excited singlet state with a lifetime of 13.5 ps [Yamazaki et al., PRL 2015] have represented the first time that EMS measurements of short lived transient species are feasible, opening the door to time-resolved orbital imaging in momentum space. With further technical development, TR-EMS could eventually enable one to take a series of snapshots of molecular orbitals changing rapidly during chemical reaction, thereby making it possible to exploit a new area for studies of ultrafast molecular dynamics as well as the nature of molecular excited states; it is electrons that bind atoms into molecules, and chemical reactions are all about the rearrangement of these electrons or the change in spatial patterns of the corresponding molecular orbitals. In this contribution, some results of our recent studies will be presented, which may examine the current status and future prospects of EMS.