Present status and future prospect of a new real-time spectroscopic complex using high-energy electron scattering

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Synopsis

We report the present status of a new real-time spectroscopic complex using high-energy electron scattering. It consists of time-resolved versions of electron and atomic momentum spectroscopies, both of which employ laser and electron pulses in a pump-probe scheme. An emphasis will be placed on two running attempts that aim to draw information about fast electron dynamics and intramolecular atomic momentum distributions.

High-energy electron scattering is one of the most useful tools to probe atomic and molecular systems. Indeed, different types of electron impact spectroscopies have been developed and applied so far to study various physical and chemical properties of matter [1]. In this regard, however, one might wonder what more there is to learn about atomic and molecular systems from the study of such high-energy electron impact spectroscopies, particularly information additional or complementary to that obtained from photon impact spectroscopies. The answer to this question is clear: one can see two good examples of this in the use of electron-impact-induced Compton scattering [2]. One example is (e,2e) electron momentum spectroscopy (EMS) [3] and the other is atomic momentum spectroscopy (AMS) [4].

EMS has a long history as an advanced form of the traditional Compton scattering experiments. The most remarkable feature of this technique is that electron momentum distribution can be measured for each electron orbital, whose binding energy is resolved. On the other hand, AMS is a quite new technique and is the complete electron analog for neutron Compton scattering. It gives momentum distributions of individual atoms with different mass numbers, involved in a system. These almost unique features of high-energy electron scattering should be made more use of for promoting and exploiting future molecular science.

With the above-mentioned motivation, we have recently developed time-resolved EMS (TR-EMS) that employs laser and high-energy-electron pulses in a pump-probe scheme [5]. The potential ability of TR-EMS has already been demonstrated such through orbital imaging of the highest occupied molecular orbital of a short-lived (13.5 ps) transient species [5] and an electronic structure study for a relatively long-lived (86 ns) molecular excited state [6]. Furthermore, we have directed our efforts also towards development of time-resolved AMS (TR-AMS). The current status of this issue is that a highly-sensitive AMS spectrometer has been constructed [7], whose signal count rate has been increased by a factor of more than 800 compared to existing setups. The goal of this project is to make a joint use of TR-EMS and TR-AMS as a new real-time spectroscopic complex and one of the challenges to be tackled is visualizing the relationship between changes of electron and atomic motions to reveal the driving force of each chemical reaction.

In this contribution, we review the present status of the new real-time spectroscopic complex. Special focus will be paid to two running attempts. One is to draw information about electron dynamics in a chemical reaction, beyond the instrumental time-resolution, from the measured TR-EMS data [5]. Our idea is that information about such fast electron dynamics may be left as a trace in the experimental data. Another is to elucidate effects of the target-gas-beam propagation direction, with respect to the momentum transfer vector, on AMS spectra. The detailed knowledge regarding effects of such target translational motion would form the basis for drawing information about intramolecular atomic momentum distributions.

References


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