

Fast-ion-induced secondary ion emission from submicron droplet surfaces

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Synopsis To investigate the complex molecular reactions induced by MeV-energy heavy ions in liquids, we have developed a new experimental system that enables mass spectrometric studies of secondary ions emitted from liquid surfaces using microdroplet targets. Coincidence measurements with forward-scattered ions provide a systematic and comprehensive analysis of the positive and negative secondary ions. In addition, the penetration length of projectile ions in droplet targets was evaluated from the amount of energy loss of the forward-scattered ions. The results highlight the unknown “submicron effects” mechanism in secondary ion emission processes.

When fast heavy ions travel through condensed matter, such as liquids and living cells, violent physicochemical reactions are caused along their trajectories. The molecular-level reaction mechanism has been studied for a long time to understand this phenomenon better. However, due to the complexity of the reaction, the details are still poorly understood. Mass spectrometry is an effective tool for comprehensively analyzing the complex fragment ions and reaction product ions produced by heavy-ion collisions. Thus, mass spectrometry of secondary ions emitted from surfaces can be a powerful probe of reactions proceeding around heavy-ion tracks. However, since mass spectrometry requires a high vacuum environment, its application to volatile liquid surfaces requires special experimental techniques.

Recently, we have developed an experimental system using microdroplets as a new approach to realize mass spectrometry on liquid surfaces[1]. In particular, coincidence measurements with forward-scattered ions have enabled systematic and quantitative analysis of positive and negative secondary ions. Furthermore, the energy loss of the forward-scattered ions provides the penetration length of the forward-scattered ions in droplet targets. Thus, the coincidence measurement provides selective information depending on the target thickness at a submicron level.

A schematic diagram of the experimental setup is shown in Fig.1. Microdroplets were generated by ultrasonic atomization under 1-atm Ar and then transported to the collision chamber with a differential pumping system. The droplets were irradiated with MeV-energy ion beams from a 2-MV Pelletron accelerator. Positive and negative secondary ions were separately analyzed by time-of-flight (TOF) mass

spectrometry using a coincidence technique in which forward-scattered ions were detected with a Si semiconductor detector. Correlations between the TOF and the energy of forward-scattered ions were recorded for each event [2].

First, we demonstrated the measurements using ethanol droplets. Positive fragment ions showed that rapid proton transfer occurs before dissociation. Unexpectedly, negative ions produced a wider variety of products than positive ions. To reveal the underlying mechanism of the formation of negative reaction product ions, we performed measurements on deuterated methanol. We proposed a molecular growth mechanism based on the association reaction between negative and neutral fragments. In addition, variations in the mass spectra and kinetic energy of secondary ions were observed in the submicron scale, although that mechanism is entirely unknown.

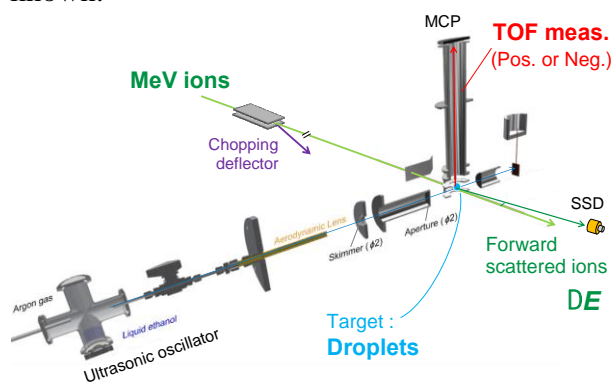


Figure 1. Schematics of the experimental setup.

References

- [1] Majima T et al. 2020 *J. Chem. Phys.* [153](#) [224201](#)
- [2] Majima T et al. 2022 *J. Phys. Chem. A* [126](#) [8988](#)

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