Soft X-ray induced ultraviolet fluorescence emission from bulk and interface of a liquid water microjet

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Synopsis Tremendous progress has been made in the research on the structure and dynamics of liquids due to the development of advanced experimental techniques such as liquid microjets, enabling investigations on volatile samples in ultrahigh vacuum environments. The spectroscopy of charged particles, e.g. photoelectron or Auger electron spectroscopy on liquids, is an established field by now. Here, we report on the successful application of a fluorescence spectrometer to measure optical emission spectra from liquids irradiated with soft X-ray synchrotron radiation.

The application of various experimental techniques to liquid samples is an ongoing challenge, especially if high vacuum conditions are required for either exciting the sample or detecting reaction products (or both). Some common tools, such as electron spectroscopy, for the investigation of molecular dynamics thus only became applicable to volatile samples with the introduction of liquid microjets [1]. With high-pressure injection of a liquid jet of a typical diameter of some tens of micrometers, sufficient vacuum conditions can be retained. Recent experiments revealed numerous processes of electronic and nuclear dynamics in liquids and solutions [1, 2]. A major remaining constraint is that the detection of electrons is a rather surface sensitive method.

Here, we report on the detection of ultraviolet fluorescence emission from the bulk of a liquid water microjet after irradiation with soft X-ray synchrotron radiation [3]. Different features appear in the emission spectrum, which can be assigned to characteristic processes. The mechanisms observed in the present work are illustrated in Figure 1. Well-known emission features from gas phase water in the jet vicinity can be found. From the liquid phase, two hitherto not reported mechanisms were observed. After inner-shell excitation of water, the Auger electron yield at the liquid–vacuum interface is that high that gas phase molecules can be excited by electron impact. This process (labelled II in Figure 1) is identified by scanning the exciting-photon energy and determining the excitation function of certain gas phase emission features [3]. For inner-shell excitation at the oxygen 1s edge, the absorption cross section is very large, leading to an immense enhancement of this surface-specific process.

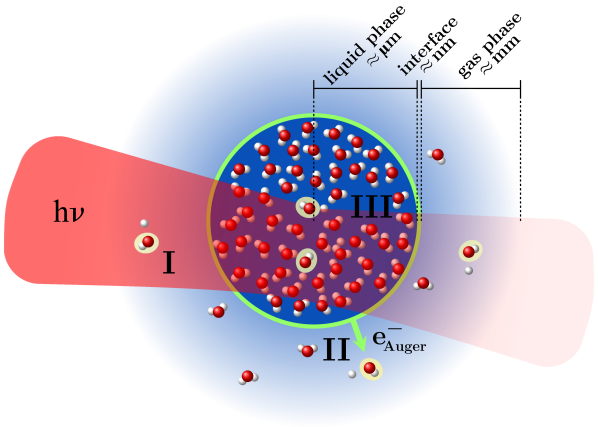

Figure 1. Illustration of radiative processes observed after soft X-ray irradiation of a liquid microjet [3].

Off-resonance, the X-rays can penetrated somewhat deeper into the jet (feature III). Here, we observe a broad genuine emission band in the ultraviolet range with a steep cutoff at the absorption edge of liquid water. Both interpretations of features II and III are supported by lifetime measurements of the respective radiative states.

References

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