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Pyridinium–Water Nanodroplets under Irradiation

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The role of impurities in water cluster growth is a crucial step in the early stage of aerosol and cloud formation [1]. In this context, the evaporation of water molecules from out-of-equilibrium pyridinium–water cluster ions was investigated using the recently developed correlated ion and neutral time-of-flight (COINTOF) mass spectrometry technique in combination with a velocity-map imaging (VMI) device [2]. This COINTOF-VMI combination allows the direct measurement of the velocity distribution of the water molecules evaporated from excited clusters after high-velocity cluster – atom collision.

Besides a low-velocity contribution, which corresponds to the statistical evaporation of water molecules after nearly complete redistribution of the excitation energy within the clusters, a high-velocity contribution is also found in which the molecules are evaporated before the energy redistribution is statistically complete. These two different evaporation modes were previously observed and interpreted for protonated water cluster ions [3]. However, unlike in the case of pure water clusters, the low-velocity part of the distributions for pyridinium-doped water clusters is itself composed of two distinct Maxwell–Boltzmann distributions (Figure 1), indicating that evaporated molecules originate in this case from out-of-equilibrium processes [4]. Statistical molecular dynamics simulations (SMD) were performed to (i) understand the effects caused in the ensuing evaporation process by the various excitation modes at different initial cluster constituents and to (ii) simulate the distributions resulting from sequential evaporations [4-5].

The evaporation of water molecules from excited clusters is found to be much slower when the cluster is doped with a pyridinium ion. Therefore, the presence of a contaminant molecule in the nascent cluster changes the energy storage and disposal in the early stages of gas-to-particle conversion, thereby leading to an increased rate of formation of water clusters, consequently facilitating homogeneous nucleation at the early stages of atmospheric aerosol formation.

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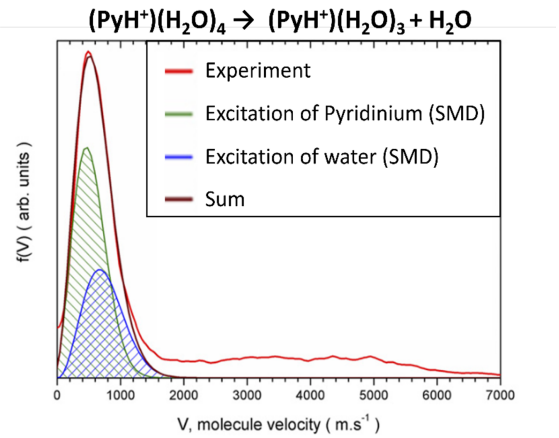

Figure 1: Measured velocity distributions of water molecules evaporated from $(PyH^+)(H_2O)_4$ (red curve) and 2 SMD calculated MB contributions corresponding to the local excitation of a water molecule (blue curve) or the pyridinium ion (green curve), whose sum is also represented (brown curve).