Abstract: Aqueous salt solutions occur widely in systems ranging from industrial processes to biological materials. Prominent examples include batteries and desalination. The properties of aqueous electrolyte solutions involve the dynamics of water and the dynamics of ions. A closely relate problem is proton transfer in acid solutions. Proton transfer in water is ubiquitous and a critical elementary event which, via proton hopping between water molecules, enables protons to diffuse much faster than other ions. While there have been a vast number of experiments and molecular dynamics simulations investigating proton hopping in water, a direct experimental observation of proton hopping has remained elusive due to its ultrafast nature and the lack of direct experimental observables. The dynamics of the formation and dissociation of complexes of Li+ and water with methylthiocyanate (MeSCN) in very concentration LiCl solutions are explicated using two dimensional infrared (2D IR) Chemical Exchange Spectroscopy. 2D IR is also used to measure the spectral diffusion, which provides information on the dynamic of the concentrated salt solutions. The slowest component is assigned to the time for ion clusters to randomize. 2D IR Chemical Exchange Spectroscopy was also used to extract the chemical exchange rates between hydronium and water in HCl solutions using MeSCN. Ab initio molecular dynamics simulations demonstrate that the chemical exchange is dominated by proton hopping. The observed experimental and simulated acid concentration dependences as well as a number of factors obtained from the simulations and spectral diffusion measurements make it possible to extrapolate the measured single step proton hopping time in concentrated HCl to the dilute limit. The results indicate that the H-bond rearrangement of the water network in which hydronium ions are embedded triggers proton hopping.