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Diffusion of hydrogen and deuterium atoms and molecular hydrogen production on amorphous solid water

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In interstellar dense clouds where a radiation field is very weak, nonenergetic reactions of hydrogen atoms on dust grains, including quantum tunneling reactions, play an important role in chemical evolution. For example, it has been widely accepted that molecular hydrogen which is the most abundant molecules in the Universe is formed by recombination of H-adatoms on the dust surface. Successive reactions of H- and D-atoms with carbon monoxide are also key molecular steps in the synthesis of many organic molecules observed in the ice mantles, such as formaldehyde and methanol.

These reactions proceed through the sequence of elementary processes of H-atoms, i.e., adsorption, diffusion, and encounter with another adsorbed species. Therefore, to understand formation of not only molecular hydrogen but also complex molecules, above-mentioned physical and chemical properties of H- and D-atoms on cold surfaces should be clarified.

In this talk, we report a spectroscopic approach for the behavior of H-atoms on the surface of amorphous solid water (ASW) using photo-stimulated desorption and resonance enhanced multiphoton ionization methods.

The diffusion rate of H-atoms was directly measured after H atom deposition on ASW at 8 K. In addition, we measured the ortho/para nuclear spin ratio (OPR) of nascent molecular hydrogen formed via recombination, and observed the spin conversion of molecular hydrogen adsorbed on ASW.

Efficient molecular hydrogen formation was observed on ASW during the H-atom deposition on ASW at 8-15 K, while some fractions of H-atoms were successfully detected even after H-atom deposition at 8 K. These results show the presence of at least two types of potential sites on ASW. The analysis of attenuation curve of H-adatoms at 8 K provides the two different activation energies of H-atom surface diffusion with about 20 meV and $>$50 meV. Quantitatively similar results were obtained in the case of deuterium atom, suggesting that the thermal hopping mechanism better explains the diffusion rather than tunneling diffusion, because a large isotope effect should be observed if it is tunneling.

The nuclear spin temperature, which is defined by OPR of hydrogen molecules, of nascent hydrogen molecules formed from H-atoms on ASW at 8 K is very close to that of adsorbed molecular hydrogen directly from the gas phase. However, when we left the hydrogen molecules on ASW, it was found to decreases on ASW by the spin conversion.

Keywords: hydrogen atom, deuterium atom, molecular hydrogen, amorphous solid water, surface diffusion