Spin conversion and catalytic reaction of hydrogen at surfaces

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The most abundant element in space, hydrogen, has unique features: Whereas hydrogen participates in a variety of chemical reactions, molecular hydrogen exists in nuclear-spin modifications according to the total-nuclear-spin multiplicity. Solid surfaces often serve as catalysts to promote nuclear-spin conversion and hydrogenation/ dehydrogenation reactions. For understanding the physics behind the processes occurring on surfaces, information on the spin and charged state of hydrogen adsorbed on solid surfaces is indispensable. Due to the limited availability of experimental techniques to probe hydrogen, however, it has been poorly understood to date. In this presentation, I discuss the role of subsurface hydrogen in a hydrogenation reaction [1,2], and magnetic/electric-field effects on nuclear-spin conversion [3,4].

In the first topic of the hydrogenation reaction, we investigated the branching of hydrogenation and isomerization of a hydrocarbon molecule on Pd nano-clusters. Figure 1 shows thermal stability of hydrogen on and in Pd nano-clusters as measured by a depth-resolved nuclear reaction technique [1]. This indicates that surface hydrogen is more stable than hydrogen absorbed in clusters. By examining the hydrogenation and isomerization of butene molecules over this Pd nano-cluster, the absorbed hydrogen was identified to participate in the hydrogenation reaction [2]. In the second topic of nuclear-spin conversion, we investigated the effects of surface magnetic field and electric field on the hydrogen nuclear-spin conversion. On an O_2-adsorbed metal surface, the spin conversion was promoted by localized spins at Ag as well as O_2 molecules [3]. On ice surfaces, on the other hand, spin conversion proceeds with a time constant of ca. 300 s (figure 2), where surface electric-fields are supposed to enhance the spin-orbit coupling (SOC) of H_2 thereby inducing the nuclear-spin flips [4].

In these phenomena, the role of subsurface hydrogen and the degree of SOC enhancement are still to be solved. For further deep understanding of spin dynamics and chemical reactions on surfaces, new experimental probes including muon and neutron are strongly required.

[1] M. Wilde et al., Phys. Rev. B 77 (2008) 113412.
[2] M. Wilde et al., Ang. Chem. Int. Ed. 47 (2008) 9289.
[3] K. Niki et al., Phys. Rev. B 79, 085408 (2009).
[4] T. Sugimoto and K. Fukutani, Nat. Phys. in press.

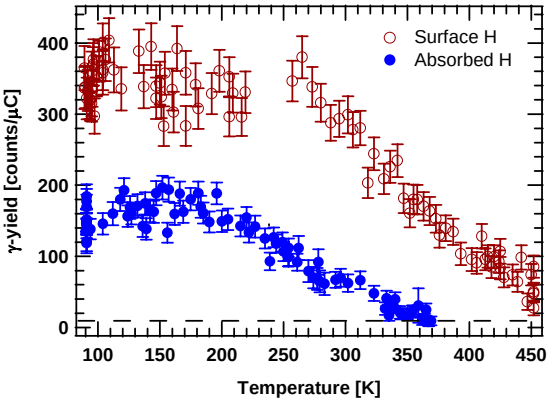

Figure 1

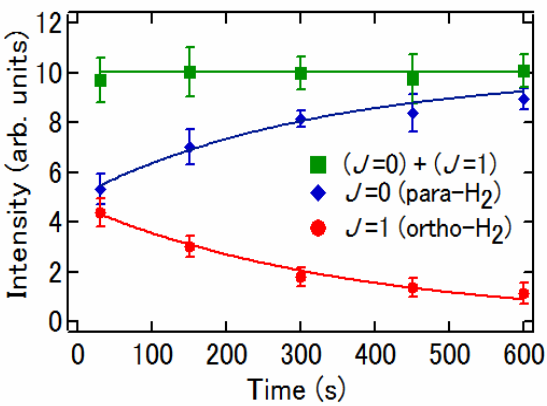

Figure 2