Energy Transfer in the Collision of H(D) Atom with Metal Surfaces and Chemical Consequences: Cu(111) vs. Pt(111)

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Energy transfer palys a key role in adsorption and reactions of gas molecules at solid surfaces. Adsorption of atoms and molecules with a typical mass is usually mediated by phonons. However, interaction of H(D) atoms with metal surfaces raises interesting questions concerning the mechanism of energy transfer; 1) despite the light mass, is phononic loss still important? 2) does electronic excitation play a significant role ? 3) is there any substrate dependence ? 4) what are the chemical consequences ? To address these fundamental points, we have measured 1) the sticking probabilities of H(D) atom, 2) the reaction cross sections for the abstraction reaction H(g) + D(ad) HD(g), and 3) the saturaction coverages of H(D) atom on Cu(111) and Pt(111) surfaces at 85 K using a pyrolytically generated H(D) atom beam of Tg = 1940 K. The sticking probabilities show a large metal dependence: s0 (H) = 0.37 on Cu(111) and 0.73 on Pt(111) at normal incidence. s0, which increases with increasing incidence angle, approaches ~ 0.52 on Cu(111) and ~ 1 on Pt(111) in the limit of zero normal energy. Large metal dependence was also observed for the abstraction reaction cross sections and the saturation coverages: σ = 5.17 2 and θs = 0.34 ML on Cu(111) and σ = 1.14 2 and θs = 0.90 ML on Pt(111).

Sticking is accompanied by the loss of normal kinetic energy, which can occur by three different mechanisms: phonic loss, electron-hole(e-h) pair creation, and normal-to-parallel momentum conversion(NPMC). First, I will discuss about the phonic loss based on a forced oscillator model to infer that phononic loss is insignificant. Next, I will argue that the transition probability of NPMC is quite small because it is a high-order diffraction. Thus, I conclude that H(D) atom loses energy on metal surfaces mainly via e-h pair generation. Then, the metal-dependent s0 can be attributed to the large difference in the density of states at the Fermi level between a sp-band metal (Cu) and a d-band metal (Pt).The large difference in the abstraction reaction can be similarily explained; the hopping range of hot H atom is larger on Cu(111) because of a less efficient energy transfer and hence a larger reaction cross section than on Pt(111). Consequently, the saturation coverage, which is determined by the competition between sticking and abstraction, is much smaller at Cu(111). Experimental results[1] on the direct measurements of electron and hole currents using Si-based Schottky diodes will be shown to corroborate my conclusion.

[1] Nienhaus et al., Phys. Rev. Lett. **82**, 446 (1999).