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Probing Excited States of Si Surfaces

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Understanding gas-surface interaction is crucial not only fundamentally, but also technologically, in areas as diverse as molecular electronics to catalysis. Yet our general understanding of the interaction has been so far limited to room temperature and above, or at a macroscopic scale, and the picture is not complete. Studies of gas-surface kinetics at low temperature and at atomic-scale may provide additional understanding of these interactions.

In this talk, we will present our studies on adsorption of oxygen and ethylene molecules on the technologically important Si(001)-c(4×2) surface using low temperature scanning tunneling microscopy. At the early stage of oxygen adsorption, reactions with Si atoms at S_B steps are dominant over those at terraces by more than 2 orders of magnitude, and they proceed in two distinct stages to high oxidation states, through the formation of -Si-O- chain structures along the step edge. On the other hand, ethylene molecules dosed at 50 K decay thermally to "di-σ" chemisorption states, which exhibits Arrhenius behavior with a small reaction barrier. Surprisingly the decay rate is found 10 orders of magnitude lower than a conventional estimate. Such a large suppression of the reaction rate is interpreted by the entropic bottleneck at the transition state induced by the free-molecule-like trap state. Our studies suggest that the free-molecule-like trap state may be quite general, including the case of oxygen molecules, and that the entropic bottleneck plays a crucial role at low temperature reactions.

In addition, we have recently developed a series of experimental techniques to apply short pulsed excitations to surfaces and probe their subsequent responses, which allowed us to study the surface dynamics at much more controlled and statistical ways. The details of techniques and the results from their applications to Si surfaces will be presented.