Contribution ID: **67** Type: **Contributed poster**

Observing the Transit CO desorption process at carbon K edge via free electron X-ray Laser

Monday, 25 June 2018 18:45 (15 minutes)

The adsorption of the gas molecule to the transition metal or noble metal surface, through a weakly bound "precursor" state which facilitate the gas molecule to lose its rotational and translation energy before forming a stable chemisorption bond, is the most fundamental elementary surface chemical process. In particular, our previous work has demonstrated the short-live transient precursor species can be "seen" by the means of oxygen resonant x-ray emission spectroscopy (XES) and x-ray absorption spectroscopy (XAS) through the pump-probe technique. It has be revealed that compared to the chemisorbed state, the CO molecules in the transient possesses a similar electronic structure to that of the gas phase, yet the anti-bonding CO 2 π^* states are still substantially influenced with the metal surface.

Here, the free-electron x-ray laser from FERMI (acronym for Free Electron laser Radiation for Multidisciplinary Investigations) has been further employed to investigate the real-time electronic structure changes at carbon K edge of CO molecules as their chemisorption state on Ru(0001) upon exciting the substrate by using a femtosecond (fs) optical laser pulse. The ultrafast pump-probe core-level excitation based on a x-ray free-electron laser provides the detailed time evolution of carbon XAS spectrum. Due to the orbital symmetry sensitivity of the oriented adsorbent, in which the linear CO molecular is perpendicularly boned to the metal surface with the carbon side facing down, the contribution of 2px, 2py, and 2pz hybridization orbitals, namely σ and π symmetry, can be resolved by regulating the E-vector polarization of incoming x-ray laser. Before the optical laser pumping, a clear broad peak centering at 288.3 eV in the horizontal E-vector spectrum should be ascribed to the CO chemisorption state. This state, yet with a relatively weak intensity, can also be seen in the vertical E-vector spectrum due to the non-ideal perpendicular adsorption of CO molecule. After the optical laser pumping, the CO precursor state with a relatively sharp peak compared to the chemisorption state starts to emerge out at 287.8 eV with the time prolonging, and meanwhile, the shifting toward the low energy of the chemisorption state peak can been observed for both polarized E-vector spectra. Our work presents, for the first time, the time-resolved observation of the CO precursor state focusing on carbon orbital level.

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Session Classification: Poster session