北京大学量子材料科学中心

International Center for Quantum Materials, PKU

Special Seminar

Nonadiabatic molecular dynamics investigations on the ultrafast charge dynamics at Interfaces

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Time: 2:00pm, July 19, 2016 (Tuesday)

2016 7 19 2:00

Venue: Room W563, Physics building, Peking University

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The ultrafast dynamics of photo-excited charge carriers plays an important in optoelectronics and solar energy conversion. Using nonadiabatic molecular dynamics simulation, we study the ultrafast charge dynamics at CH_3OH/TiO_2 and MoS_2/WS_2 interfaces. For CH_3OH, we study the forward and backward hole transfer between TiO_2 and CH_3OH as well as the whole hole relaxation process to valance band maximum (VBM). First, we found that the hole trapping ability of CH_3OH depends on the adsorption structure strongly. Only when the CH_3OH is deprotonated to form chemisorbed CH_3O, there will be ~15% hole trapped by the molecule. Second, we found the time scales of forward hole transfer process from TiO_2 to CH_3O (hole trapping process) and hole relaxation to VBM strongly depend on the temperature. When the temperature decreases from 300K to 30K, for hole trapping process, the time scale increases from 150 fs to ps magnitude. The hole relaxation process to VBM is also slow down significantly. This can be interpreted by the reduction of the non-adiabatic coupling and the phonon occupation. Our studies provide valuable insights into the photogenerated charge dynamics near molecule/TiO_2 interface.

For MoS_2/WS_2, we show that instead of direct tunneling, the ultrafast interlayer hole transfer is strongly promoted by an adiabatic mechanism through phonon excitation. At room temperature the interlayer charge transfer in MoS_2/WS_2 is ultrafast with a timescale of 20 fs which is in good agreement with the experiment. This ultrafast hole transfer process can be suppressed by decreasing the temperature to 100K, which reduces the phonon occupation and the charge transfer is then dominated by direct tunneling, which happens at the time scale longer than 300 fs. The atomic level picture of phonon-assisted ultrafast mechanism revealed in our study is valuable both for the fundamental understanding of ultrafast charge carrier dynamics at vdW hetero-interfaces as well as for the design of novel quasi-2D devices for optoelectronic and photovoltaic applications.

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