Dynamics of chemical reactions at surfaces under thermal excitation by laser irradiation and probed by time resolved SFG spectroscopy

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The dynamics of surface molecules with ps time resolution gives detailed information for elemental steps of chemical reactions at surfaces such as catalytic reactions. TiO_2 surfaces have been utilized to catalysts and photocatalysts. The time resolved SFG spectroscopy with NIR pump pulses reveals the ultra-short dynamics of structural change of formate species on TiO_x. The temperature jump at TiO_x layers prepared on a Pt(111) surface was initiated by the 1064 nm pulses (35 ps pulse width) and the temperature was relaxed to the initial temperature within ns time scale. The surface species (deuterated formates) changed the structure during the temperature change, which is hard to be observed by conventional static spectroscopy. The thermal broadening of vibrational peaks and population changes between different surface species showed the unique changes of transient SFG signals [1.2].

Photocatalytic water splitting is one of the powerful tools to convert solar energy to solar hydrogen. We performed in-situ RAIRS experiments to understand the catalytic selectivity and the potential changes at photocatalyst surfaces [3,4]. This system is promised to be a valuable subject for the ultra-fast vibrational spectroscopy.

[1] J. Chen, et al., *J. Phys. Chem.*, **112**, 12477 (2008)
[2] J. Chen, et al., *J. Am. Chem. Soc.*, **131**, 4580 (2009)
[3] M. Yoshida, et al., *J.Phys. Chem.*, **113**, 10151 (2009)
[4] M. Yoshida, et al., *J. Am. Chem. Soc.*, **131**, 13218 (2009)

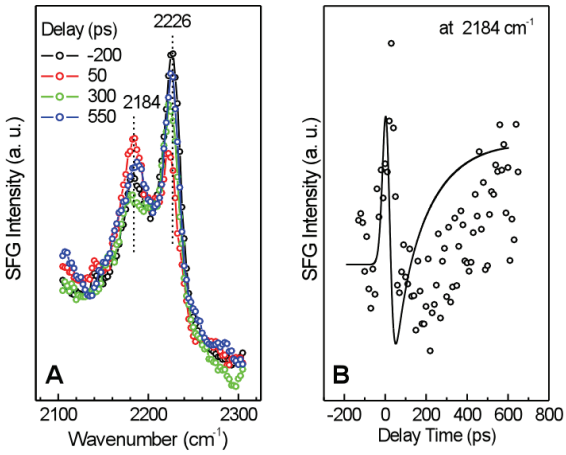

Figure 1: Transient SFG spectra (A) and signal change at 2184 cm^{-1} (B) of *d*-formates on TiO_x/Pt(111) at 250 K under the irradiation with 1064 nm pulses [2].