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In-situ observation of water formation reaction on Pt(111) with dispersive NEXAFS

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Introduction

The catalytic formation of water from adsorbed oxygen and hydrogen atoms on Pt(111) has been extensively studied because it is one of the simplest model reactions to understand the mechanisms of heterogeneous catalyses at the atomic level. Although a recent STM study [1] has provided a deep insight into the reaction mechanism, there still remains open questions, particularly about behaviour of an intermediate species (OH). This is because STM observations give only information on a local area of a microscopic scale, while the two-dimensional distribution of the intermediate species rapidly propagates in a mesoscopic scale. It is essential for the understanding of this reaction to observe real-time change of the reaction species on the whole surface. Dispersive NEXAFS [2] is a powerful tool to conduct the observations of the reaction with spectroscopic approach. Here we report preliminary results of dispersive NEXAFS measurements for the water formation reaction on Pt(111).

Experimental

The experiments were performed at BL-7A using an ultrahigh vacuum system (2.0×10^{-10} Torr). This system is equipped with a position-sensitive electron energy analyzer which is used for detecting Auger electrons emitted from the surface. Since the sample surface was irradiated by energy dispersed x rays, the position-sensitive detection of the Auger electrons provides a NEXAFS spectrum by Auger electron yield in a one-shot manner. It takes usually 40 sec to obtain a spectrum with reasonably high signal-to-noise ratio.

The Pt(111) surface was pre-covered by a p(2x2) layer of atomic oxygen. This surface was exposed to gaseous hydrogen ($0.1\text{-}1.0\times10^{-8}$ Torr) at a typical surface temperature of 130 K, where no water desorption takes place. Dispersive O K-edge NEXAFS spectra were taken during hydrogen exposure.

We also measured NEXAFS spectra of an OH-covered Pt(111) surface by the conventional partial-electron-yield method as the standard. The OH-covered surface was prepared by dosing H_2O on (2×2)-O/Pt(111) at 135 K and subsequent annealing at 175 K.

Results and discussion

Typical results of dispersive-NEXAFS measurements at normal incidence are shown in Fig.1. Although it is obviously recognized that a peak due to atomic oxygen (530 eV) disappears and a broad structure mainly associated with water (540 eV) increases with exposure, no OH-induced peak appears during the reaction. The absence of OH feature is due to a small transition probability of the OH species for the normal-incidence spectra at the low-energy region as shown in Fig. 2. The OH species gives a strong peak at 531 eV in the grazing-incidence spectrum. Therefore we measured dispersive NEXAFS at grazing incidence and found a component that is ascribed to the OH species after properly subtracting the atomic-oxygen component. Time-dependent change of the surface O, OH and H_2O species during the water-formation reaction has been thus obtained and kinetic analyses for the data are now underway.

References

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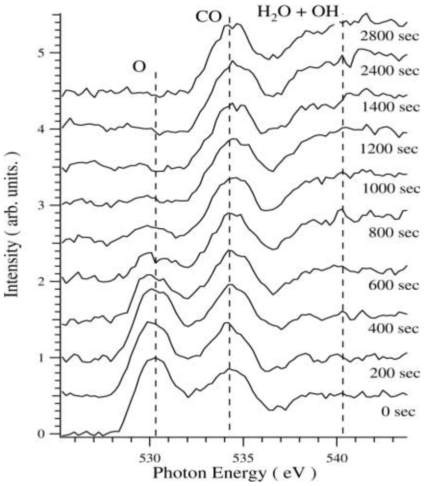

Fig. 1. Dispersive NEXAFS at normal incidence (150 K, hydrogen pressure 8.0×10^{-9} Torr).

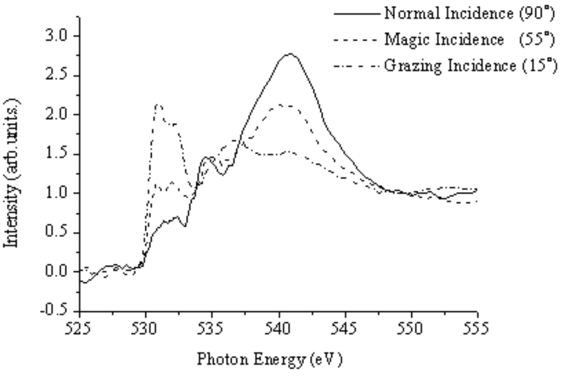

Fig.2 . NEXAFS of OH – Pt(111) at 150 K