

論文の内容の要旨

論文題目 Interface structures of metal nanoparticles and oxide surfaces
 (酸化物を担体とする金属ナノ粒子の界面構造)

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In the present thesis, we employ aberration-corrected STEM to perform systematic studies of the interaction between oxide and metal nanoparticles. Pt/TiO₂ and Au/TiO₂ are chosen as the target system because although they have been extensively studied due to their importance, little is known about the role of the surface chemistry, the interaction between support and nanoparticles, as well as the growth behavior of the nanoparticles, in particular in atomic dimension. Therefore, we performed intensive investigations on two dimensions: single atom and nanoparticle study, looking forward to elucidating the catalytic performance of heterogeneous catalysts, and also developing a method that we can apply to other nanocluster/supporting materials study.

In chapter 2, we demonstrated the support effects of TiO₂ (110) surface on the early stage of Pt nanoparticle growth by evaporating Pt nanoclusters onto amorphous carbon and TiO₂ (110) surface, using HAADF-STEM images to analyze the size, shape, and the intensity distribution of Pt nanoparticles on both substrates and comparing their differences. In chapter 3, we study the preferential adsorption sites of Pt atoms on TiO₂ (110) surface by aberration-corrected STEM and first-principles calculation. Surface oxygen vacancies, especially basal oxygen vacancy, serve as the most stable for Pt single atom adsorption. In chapter 4, we evaporate different amount of Pt onto the TiO₂ (110) surface and use HAADF-STEM to characterize their atomic structures. Pt was found to form small nucleus on the surface of TiO₂ (110) around surface oxygen vacancies, which are the most likely sites to anchor Pt atoms. As Pt becomes larger nanoclusters, the interface loses lattice coherency to accommodate larger lattice mismatch between Pt and TiO₂. In chapter 5, we employ the method we developed in previous chapters to study the interface structure of Au/TiO₂ (110). It is found that surface oxygen vacancies also governs the adsorption of Au atoms on TiO₂ (110) surface. However, unlike Pt atoms are found on all the possible surface sites and mostly on basal oxygen vacancies, Au atoms only attach to bridging oxygen vacancies. DFT calculations suggest this difference may result from the ground electronic state of Au and Pt. Chapter 6 presents a summary of this thesis.