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STEM study of Au nano-particles supported on metal oxides

Tomoki Akita[1,3], Shingo Tanaka [1], Koji Tanaka [1], Masatake Haruta [2,3] , Masanori Kohyama [1,3]

[1] Research Institute for Ubiquitous Energy Devices, National Institute of Advanced Industrial Science and Technology (AIST), Midorigaoka 1-8-31, Ikeda, Osaka 563-8577, Japan
[2] Department of Applied Chemistry, Graduate School of Urban Environmental Sciences, Tokyo Metropolitan University, Minami-Osawa1-1, Hachioji, Tokyo192-0397, Japan
[3]JST-CREST

It is well-known that the Au nano-particles supported on metal oxides exhibit high catalytic activity [1]. It was also pointed out that the catalytic properties of Au nano-particles are sensitive to the size of Au particles and the interface structure between Au and metal oxide support [1,2]. Some experimental results indicate that the perimeter of Au particle and metal oxide interface plays key role for the low temperature CO oxidation, but the details are not understood. Thus, it is important to elucidate the structure of Au nano-particles and metal oxides interface in atomic scale by electron microscopy.

In this study, the structures of Au particles on TiO_2 and CeO_2 were observed by an analytical transmission electron microscopy (TEM), JEOL JEM-3000F, equipped with annular dark field scanning transmission electron microscopy (HAADF-STEM) systems. The Au/CeO_2 and Au/TiO_2 model catalysts were prepared by using the substrates of CeO_2 poly-crystal and TiO_2 single crystal. The Au nano-particles were deposited on the metal oxide substrates by a conventional vacuum deposition. The Au particles of 2-5 nm in diameter were deposited on the substrates by controlling the amount of Au. The interface structure was also observed by HRTEM. The orientation relationship of (111)[1-10]Au//(111)[1-10]CeO_2 was frequently observed in profile-view HRTEM images for Au/CeO_2 samples [3,4] while the obvious preferential orientation relationships were not observed for Au particles on TiO_2. High resolution HAADF-STEM images were also obtained for Au-TiO_2 and Au-CeO_2 interface as shown in figures 1 and 2. The position of atomic columns of Au, Ti and Ce at interfaces is directly investigated from HAADF-STEM images while the oxygen columns are not detected in the HAADF-STEM images. The distance between Au and metal oxide supports was estimated from the intensity profile of HAADF-STEM images as 0.33nm and 0.28nm for Au-TiO_2 and Au-CeO_2, respectively. The interface structure between Au-Ti and Au-Ce were discussed by comparison with the stable structure of the interfaces obtained from first principle calculations considering the oxygen defects of the oxide surface. The structure changes are also observed by HAADF-STEM for the Au particle on CeO_2 surface. The hemispherical Au particle changed to fat shape during electron beam irradiation. The atomic columns of Au were observed by HAADF-STEM until the height of Au particle changed to 2 atomic layers. The flat Au particle changes to hemispherical shape again without electron beam irradiation. The disordered structure was also observed at the perimeter interface between the Au particle and CeO_2 substrate.

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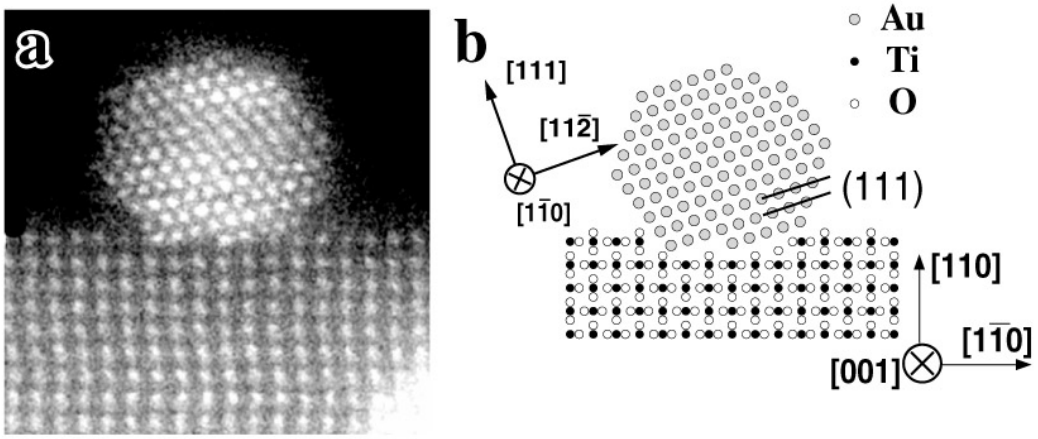

FIG. 1. HAADF-STEM image of Au particle on TiO_2(110) surface.

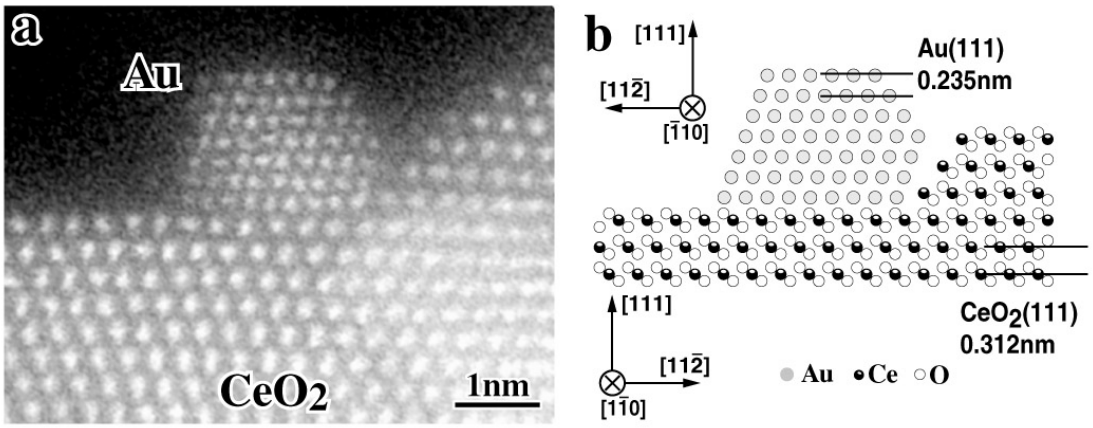

FIG. 2. HAADF-STEM image of Au particle on CeO_2(111) surface.