Gold-Assisted Growth of Pyramid-shape SrTiO₃ Bases During Thermal Dewetting

Anchao Yi¹, Fang Liu¹, Ming-hui Lin¹ and Guo-zhen Zhu^{2,1}

Gold-oxide nanostructures have attracted extensive attention because of their widespread applications in biomedicine and catalysis [1-2], and the corresponding properties are sensitive to the gold-oxide interface and/or nanoparticle morphology [2-4]. Recently, we reported the reconstruction of gold-spinel (MgAl₂O₄) interfaces, which have comparably low interface energy. In relation to the special interface structure, oxide bases grow underneath gold nanoparticles from nominally stable oxide substrate [5-6]. The clarification of the interface structure and this new phenomenon can be significant to extend our understanding of gold-oxide nanostructures.

Herein, we study a similar lattice-matched system, Au-SrTiO₃ (4.078Å for Au and 3.905Å for SrTiO₃). In order to facilitate the following structural characterization, we prepare gold nanoparticles on the surface of single-crystal (111) SrTiO₃ substrates by dewetting, which involves the sputtering of gold film on the substrates, followed by the heat treatment with the temperature of 800°C, 900°C, 1000°C and 1100°C, respectively. As shown in Fig.1(a), the initial gold film breaks into nanoparticles after the heat treatment, and the particle radius varies with the initial film thicknesses and the heat-treatment temperatures. Facets of the dewetted particles are detected. Some particles have grooves, which is related to the formation of twins. The facets and grooves of small particles are nearly invisible probably due to the limited resolution of the microscope. More importantly, as shown in the white arrows in Fig.1(a) and the HAADF image in Fig.1(b), when the heat treatment temperatures are relatively high (e.g. 1000°C and 1100°C), the oxide bases grow underneath these dewetted gold particles with a height of about 10-30nm.

The crystallographic orientations of gold particles are studied by a X-ray polycrystalline diffractometer. According to the generated $\{111\}_{Au}$ pole figures (Fig.2), the $\{111\}_{Au}$ planes of particles prefer to parallel the substrate surface and their in-plane orientations are randomly at lower temperatures. After the heat treatment at 1100° C, there are one main orientation: $\{111\}_{Au}/\{111\}_{SrTiO3}\&\langle 1\bar{1}0\rangle_{Au}/\langle 1\bar{1}0\rangle_{SrTiO3}$ and its twinned orientations: $\{115\}_{Au}/\{111\}_{SrTiO3}\&\langle 1\bar{1}0\rangle_{Au}/\langle 1\bar{1}0\rangle_{SrTiO3}$ as well as $\{11111\}_{Au}/\{111\}_{SrTiO3}\&\langle 1\bar{1}0\rangle_{Au}/\langle 1\bar{1}0\rangle_{SrTiO}$. These preferential orientations are the same in the previously reported case of Au-MgAl₂O₄ system [7].

The interface structure between nanoparticle and grown bases is examined by scanning electron transmission microscopy (STEM). The grown bases have the same lattice features and are epitaxial to the substrate. Gold {111} planes are directly connected to the topmost Ti monolayer of SrTiO₃ lattice, without noticeable chemical or structural changes. The inter-planar spacing between this gold monolayer and Ti monolayer is measured to be ~2.0Å, which is slightly smaller than the inter-planar spacing of {111} Au planes (2.35 Å). Further study is carrying out about the effect of interface and its energy on the grown bases [8].

References:

^{1.} State Key Laboratory of Metal Matrix Composites, School of Materials Science and Engineering, Shanghai Jiao Tong University, Shanghai, P.R. China.

^{2.} Department of Mechanical Engineering and Manitoba Institute for Materials, University of Manitoba, Winnipeg, MB, Canada.

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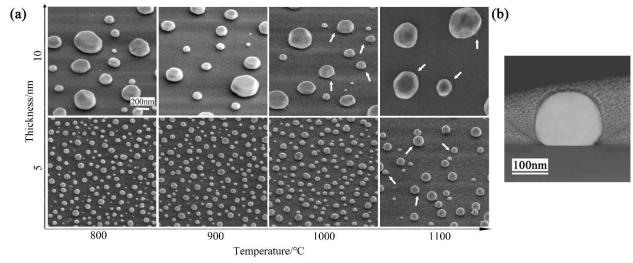


Figure 1. The morphology of gold nanoparticles. (a) The SEM micrograph of nanoparticles on the substrates. (b) The HAADF (Z-contrast) image presenting the cross-sectional view of nanoparticle on grown bases.

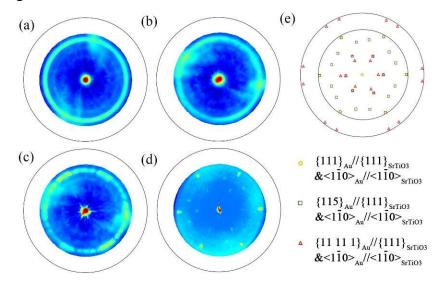


Figure 2. The $\{111\}_{Au}$ pole figures with heat treatment temperatures of (a) 800°C, (b) 900°C, (c) 1000°C and (d) 1100°C. (e) is the simulated pole figure from possible orientations.

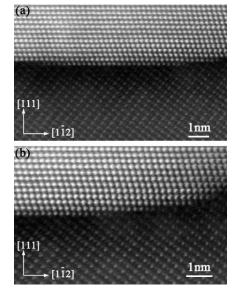


Figure 3. The STEM-HAADF images of Au-SrTiO₃ interface.