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Selective-area-growth of rutile-type SnO_2 on TiO_2 (110) substrate

Selective-area growth to produce high-quality rutile-type oxide semiconductors for the development of next-generation power devices

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Summary

- **The growth mechanism of rutile-type tin oxide (r-SnO_2), one of the rutile-type oxide semiconductor candidates for next-generation power devices, was investigated in detail using electron microscopy.**

- **The dislocation-propagation behavior was clarified, and guidelines for the fabrication of high-quality rutile-type oxide thin films with low dislocation densities were established.**

Hitoshi Takane, a Ph.D student of Kyoto University, and co-researchers have clarified the characteristics of selective-area growth mechanism used to produce higher-quality rutile-type oxide semiconductors. In an experimental study of growing a rutile-type tin oxide (r-SnO_2) on a rutile-type titanium oxide (r-TiO_2) substrate, the authors succeeded in clarifying the growth dynamics and forming regions with few defects (dislocation-free regions). In the future, this research will be applied to other rutile-type oxide semiconductors to produce high-quality thin films required for the development of next-generation power devices.

Detail

Realization of rutile-type oxide semiconductors with fewer defects

Power semiconductors are used in everything from commonplace smart devices, such as personal computers, to power circuits in electric vehicles. Development of next-generation power semiconductor materials is currently underway. In particular, research and development of "ultra-wide bandgap (UWBG) semiconductors," which have an extremely wide bandgap (*1) and are directly linked to the development of innovative power devices, is accelerating. UWBG semiconductor candidates include gallium oxide (Ga_2O_3, band gap of 4.4 – 5.6 eV), diamond (5.5 eV), aluminum nitride (AlN, 6.0 eV), and cubic boron nitride (c-BN, 6.3 eV).

Hitoshi Takane, a Ph.D student of Kyoto University, and co-researchers focused on rutile-type oxide semiconductors, particularly rutile-type germanium oxide (r-GeO_2). r-GeO_2 is an UWBG semiconductor with a direct bandgap of 4.7 eV and characterized by its both p- and n-type dopabilities proven via theoretical calculations. The research group has successfully achieved epitaxial growth of r-GeO_2 on rutile-type titanium oxide (r-TiO_2) single-crystal substrates, which are commercially available, and has proposed a semiconductor alloy system with other rutile-type oxides. Moreover, they have already succeeded in fabrication of alloy films with rutile-type tin oxide (r-SnO_2), which has the next largest band gap of 3.6 eV after r-GeO_2, over the entire composition range.

Rutile-type oxide semiconductors exhibit large bandgaps and high functionality; however, determining their growth dynamics remains challenging. r-GeO_2 has been found to contain dislocations when epitaxially grown on r-TiO_2 substrates.

From the viewpoint of power device applications, these dislocations are known to degrade the device performance (low electron and hole mobility, increased leakage current, and premature breakdown). This study aims to reduce these dislocations and provide knowledge that will lead to the development of high-performance next-generation power devices.

In this study, r-SnO_2 was grown on a r-TiO_2 substrate with a SiO_2 mask and the microstructure was observed in detail using two different electron microscopic techniques to clarify the crystal growth mechanism and determine the conditions for realizing low-dislocation regions. In the future, the research group plans to apply this research to other rutile-type oxide semiconductors, such as r-GeO_2, to achieve high crystal quality, which is necessary for developing next-generation power devices.

Clarifying the growth mechanism of rutile-type tin oxide (r-SnO_2)

In this study, to clarify the growth mechanism of r-SnO_2, it was grown on substrates with microfabricated circular and stripe-shaped masks. The r-SnO_2 selectively grown on window areas of the masks was examined using scanning electron microscopy (SEM). The "mist CVD method" was used as the growth technique. This method has been used by various research teams to grow oxide thin films, like Ga_2O_3, zinc oxide, and so on. The team chose this method because it is simpler and less expensive than more common growth methods, including metal–organic chemical vapor deposition (MOCVD).

First, it was confirmed that the crystal island grew on the circular mask pattern by forming a hexagonal facet structure (Figure 1). The experiment revealed that r-SnO_2 crystals grow with a facet structure that reflects the “equilibrium form” (*2) of rutile-type structure based on the surface energy density derived from theoretical calculations, which is crucial for controlling the crystal growth in the future.

In the stripe-shaped mask pattern, during the initial growth stage, several small crystal islands were formed and grew three-dimensionally (Volmer-Weber mode) on the window area. Then, they coalesced each other to form a striped crystal. Finally, it was found that lateral overgrowth started after the complete coverage of the window (Figure 2).

Based on these observations, the research group was able to assess the growth process of r-SnO_2 from the initial stage. Subsequently, they determined that the facet shape of the striped sidewalls reflects the order of the surface energy density derived from the theoretical calculations, even when grown on a stripe-shaped mask pattern.

Regarding research on techniques to control low-dislocation regions

The group also observed the grown r-SnO_2 using cross-sectional transmission electron microscopy (TEM). Dislocations in the laterally grown region propagated perpendicularly to the inclined facet plane (the image force effect) (Figure 3). The formation of dislocation-free regions (present in the lower part of the laterally grown region on the mask) was also observed (dislocation density $\leq 5 \times 10^8$ cm^{-2}), confirming the effectiveness of the selective-area growth.

Although the dislocation-free region remains small, the present results indicate that facet surfaces follow an equilibrium shape that reflects the surface energy density derived from theoretical calculations. This suggests that by appropriately selecting the substrate surface orientation and mask orientation, the facet shape and dislocation behavior can be controlled.

A method for generating the dislocation-free regions is important for the mass production of power devices using rutile-type oxide semiconductors. In the future, the research group will investigate and develop methods for the epitaxial growth with even larger dislocation-free regions by appropriately selecting the substrate orientation and mask pattern shape to improve the precision of microfabrication.

Annotation

*1 **Bandgap** It denotes the energy width of the forbidden band in the band structure of a crystal. Depending on the bandgap energy, the material can be categorized as a metal, insulator, or semiconductor.
*2 **Equilibrium form** The form for which interfacial free energy in a crystal is minimized.

Figures

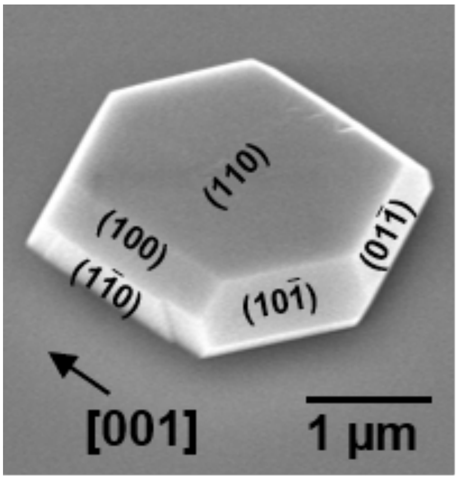

Figure 1. SEM image of r-SnO_2 single crystal selectively grown on a circular window.

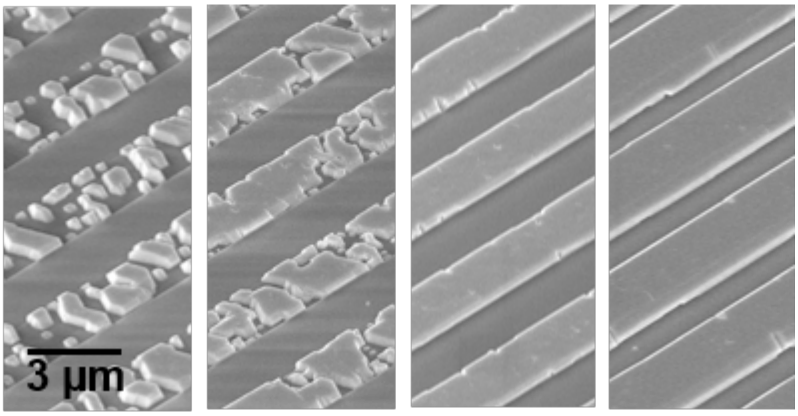

Figure 2. SEM images demonstrating the growth process of r-SnO_2 on the striped window (left to right).

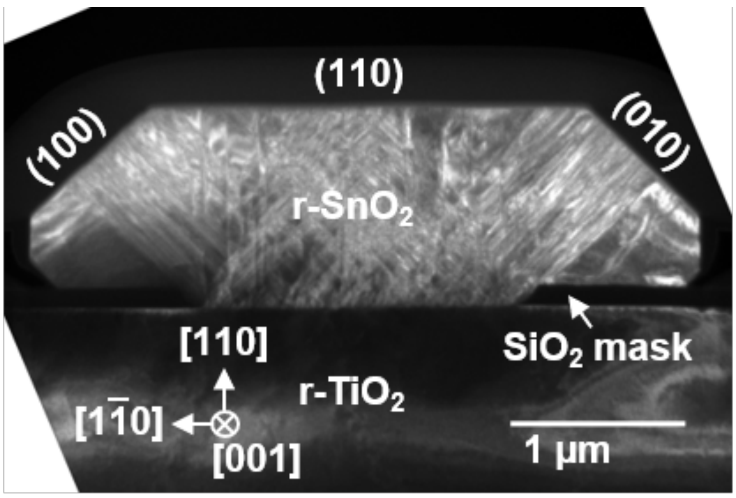

Figure 3. Cross-sectional TEM image of r-SnO_2 selectively grown on the striped window.