COHERENT THERMAL CONDUCTION IN TITANIUM-CHROME OXIDE
NATURAL SUPERLATTICES WITH AN ORDERED ARRANGEMENT OF
PLANAR FAULTS

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1. INTRODUCTION
The control of heat transport through the manipulation of phonons as coherent waves in solids by the nanoscale periodic structure is of great interested and expected to be applied advanced thermal management [1,2]. Superlattices are the ideal model systems for the realization and understanding of coherent phonon effects on macroscopic thermal properties and some researchers have reported the coherent heat conduction in superlattices [3-5]. However, it is still challenging to prepare smooth interfaces for phonons with terahertz order contributing to heat conduction at room temperature, since the interfaces in artificial superlattices are mainly incoherent. Therefore, we are focusing on the titanium oxide natural superlattice, which has nanoscale periodic arrangement of planar faults called crystallographic shear planes as a thermodynamically stable phase [6-10]. In this study, we investigated the structure and the temperature dependence of thermal conductivity for the natural superlattice.

2. EXPERIMENTAL
High-purity TiO2 and Cr2O3 powders weighted with 24.1wt%Cr2O3 were mixed and pressed into pellets. Then, the pellets of mixed powders were sintered at 1423 K for 24 h in air. Polycrystalline specimens were prepared by arc-melting the sintered pellets. The polycrystals were annealed at 1673 K in air. The structure of the crystals was evaluated by X-ray diffraction (XRD), transmission electron microscopy (TEM) and a scanning transmission electron microscope (STEM) with high-angle annular dark-field (HAADF) detector. The thermal conductivity was measured from 10 K to room temperature by static method.

3. RESULT AND DISCUSSION
Figure 1(a) shows the HAADF-STEM images taken from the prepared specimens along the [11-10]rutile zone axis. In the HAADF-STEM images, Ti/Cr atom columns are imaged as bright spots, while O atom columns are not imaged. The prepared crystal included planar faults in a periodic arrangement at intervals of 1.3 nm. Figure 1(b) shows the magnified HAADF-STEM images taken from the prepared specimens. The planar faults are composed of alternative and random placement of (121)rutile and (011)rutile. The interface roughness was evaluated to be as small as about 14 pm [10]. Figure 1(c) shows the temperature dependence of the thermal conductivity of the titanium dioxide (TiO2) polycrystal and titanium-chromium oxide natural superlattices. The thermal conductivity of natural superlattice was lower than that of TiO2, and the difference was larger at low temperatures. In order to consider the coherent thermal conduction in natural superlattice, the thermal conductivity was calculated using the Debye-Callaway model [11]. In the present calculation model, the relaxation time of planar faults scattering, boundary scattering, impurity scattering, and Umklapp scattering was considered. The relaxation time of planar faults is calculated from the following equation considering the specularity of phonons at the planar faults [5,12].

\[ \tau_f^{-1} = \frac{\nu}{L_f} (1 - p), \quad p = \exp\left(-16\pi^2 \frac{R_f}{v} \right) \]

where \( \tau_f, f, L_f, R, \nu \) and \( p \) stand for the relaxation time of planar faults scattering, frequency, period of planar faults, roughness of the interface, the group velocity of phonons and specularity parameter respectively.

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Calculated thermal conductivity with \( p = 0 \) corresponding to completely incoherent scattering at planar faults is much lower than the measured value. Here, we calculated the values of thermal conductivity using the interface roughness evaluated from STEM observation and found that the calculated thermal conductivity well reproduced the measured values. The present results indicates that the planar faults in the natural superlattice have pico-scale structural perfection and behave as coherent interface to thermal phonons both from the structural analysis and measurement of thermal conductivity.

**4. CONCLUSIONS**

A polycrystalline specimen of titanium oxide natural superlattice with an ordered arrangement of planar faults was prepared, and the structural analysis and the measurement of thermal conductivity indicates that the planar faults in the natural superlattice have pico-scale structural perfection and behave as coherent interface to thermal phonons.

**REFERENCES**


