

フォノン-ガラス電子-クリスタル特性の Magnéli タングステン酸化物

Phonon-Glass Electron-Crystal behavior in Magnéli tungsten oxide

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Phonon-Glass Electron-Crystal (PGEC, Slack 1995) is well-known concept to improve the figure of merit of thermoelectrical materials. In order to realize the PGEC concept, exotic materials satisfying very short phonon mean free path and very long electron mean free path simultaneously are required. Although several exotic materials have been found in several cage compounds like Skutterudites and Clathrates, it has not been reported among the simple composition oxides.

Here we report that oxygen reduced tungsten oxide ($\text{WO}_{3-\delta}$), so-called Magnéli, epitaxial films show very high electrical conductivity ($\sim 10^3 \text{ S cm}^{-1}$) and very low thermal conductivity ($1.7 \text{ W m}^{-1} \text{ K}^{-1}$) simultaneously. We fabricated $\text{WO}_{3-\delta}$ epitaxial films on (001) LaAlO_3 substrate under precisely controlled oxygen partial pressure by PLD. The Magnéli phase was epitaxially stabilized on the substrate and the concentration of the Magnéli phase was successfully modulated by the oxygen atmosphere during the film growth. The oxygen composition $3-\delta$ in $\text{WO}_{3-\delta}$ was modulated from 2.79 to 2.98. The electrical conductivity σ of $3-\delta \approx 3$ was less than $10^{-1} \text{ S cm}^{-1}$, and the thermal conductivity κ was $6.8 \text{ W m}^{-1} \text{ K}^{-1}$. The σ of the epitaxial $\text{WO}_{3-\delta}$ films dramatically increased with decreasing $3-\delta$ and reached 10^3 S cm^{-1} when $3-\delta \sim 2.79$. On the other hand, the κ dramatically decreased below $1.7 \text{ W m}^{-1} \text{ K}^{-1}$, which is close to that of amorphous film. The phonon mean free path of $\text{WO}_{2.79}$ was only 0.2 nm whereas the electron mean free path was 1.9 nm , indicating PGEC is realized in the Magnéli $\text{WO}_{2.79}$.

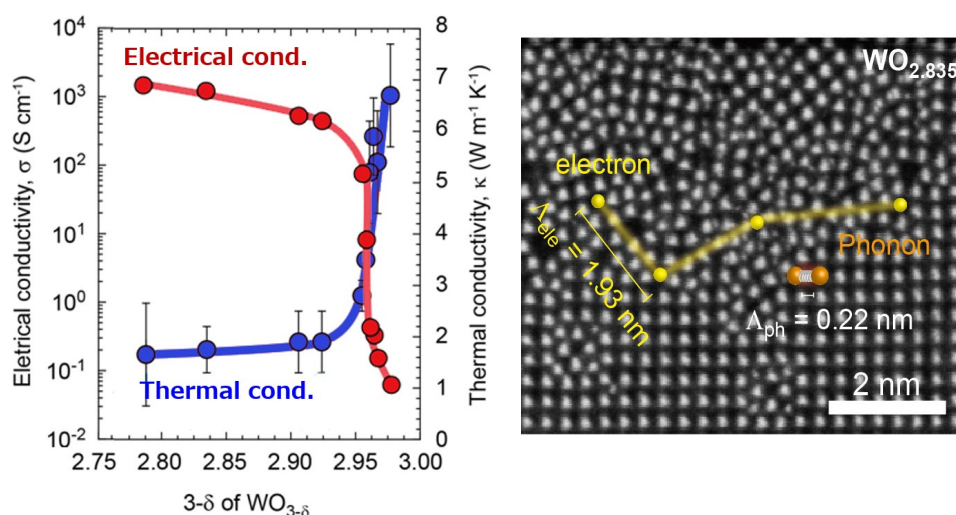


FIG. (left) Electrical and (red) thermal conductivity (blue) of the $\text{WO}_{3-\delta}$ films. Note that the electrical conductivity increases while the thermal conductivity decreases dramatically with decreasing $3-\delta$. This anomalous thermal conductivity is due to the complicated crystal structure of Magnéli $\text{WO}_{3-\delta}$ (right).