Crystallographic Anisotropy in Physical Properties of Ta_3N_5 Epitaxial Thin Films

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[Introduction] Hydrogen generation by photocatalytic overall water splitting has great potential in solving environmental problems resulted from the overuse of fossil fuels. Ta_3N_5 is a promising material that has nearly ideal bandgap (2.1eV) and band-edge positions for photocatalytic water splitting.[1] Because of its pseudo-brookite crystal structure (Fig. 1), Ta_3N_5 has been predicted to have large crystallographic anisotropy of physical properties such as band gap and carrier effective mass, which should affect its photocatalytic efficiency.[2] However, it was hard to verify these anisotropies experimentally because Ta_3N_5 has been synthesized only in polycrystalline and rod-like nanocrystalline forms so far.[3, 4] In this study, we unprecedentedly synthesized epitaxial thin films of Ta_3N_5 and investigated crystallographic anisotropy in their physical properties.

[Experiments] Ta_3N_5 thin films were synthesized by solid phase epitaxy on $LaAlO_3$ (LAO) (110) and LSAT (110) substrates, as follows. First, amorphous Ta_3N_5 films with stoichiometric chemical composition were deposited by reactive rf magnetron sputtering using a ceramic TaN target and mixture of Ar and N_2 gas. Then, the amorphous films were heated under N_2 or NH_3 flow in a tubular furnace with temperatures (T_a) ranging from 500 to 800 °C. Crystallinity, chemical composition and optical property of the films were examined by X-ray diffraction (XRD), elastic recoil detection analysis, and UV-visible spectroscopy, respectively.

[Results] XRD measurements revealed that the films annealed under N_2 showed no XRD peaks from Ta_3N_5 while the amorphous films annealed under NH_3 flow epitaxially crystallized into (010)-oriented Ta_3N_5 at $T_a \geq 700$ °C (Fig. 2). Chemical composition of the epitaxial Ta_3N_5 films was almost stoichiometric (N/Ta = 1.63). From these results, we concluded that epitaxial thin films of Ta_3N_5 were obtained. Optical reflection spectra of the epitaxial Ta_3N_5 thin films evaluated with linearly polarized light showed clear anisotropy in absorption edge and obvious difference in film's color was observed (Fig. 3), which agreed well with theoretical prediction.[2] Physical properties of the epitaxial Ta_3N_5 thin films will be further discussed in the presentation.

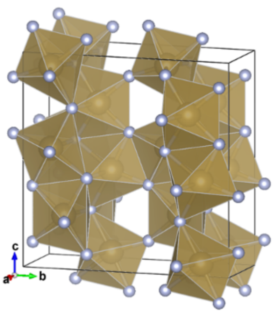

Fig. 1. Crystal structure of Ta_3N_5.

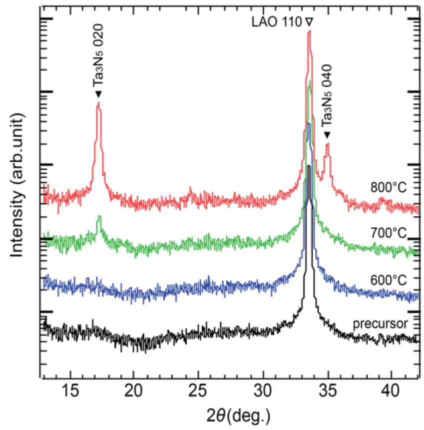

Fig. 2. XRD patterns of the Ta_3N_5/LAO films annealed for 1 hour under NH_3 flow at various T_a.

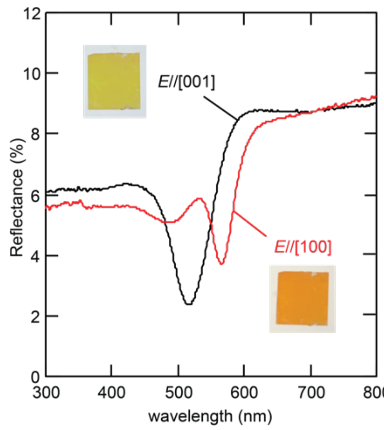

Fig. 3. Polarized optical reflectance spectra and photographs of a Ta_3N_5 epitaxial thin film.

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