低濃度アルミニウムドープされた単結晶 $SrTiO_3$ 薄膜の光触媒活性

Photocatalytic activity of low-level Al-doped single crystal $SrTiO_3$ thin films

○(D)馬 嘉悦 [1]，リップマー ミック [1] (1. 東大物性研)

°(D)**Jiayue Ma[1], Mikk Lippmaa[1](1. ISSP, Univ. of Tokyo)**

E-mail: mjiayue@issp.u-tokyo.ac.jp

The photoelectrochemical (PEC) activity of semiconductor water splitting photocatalysts is a direct and convenient measure of the light-to-fuel energy conversion efficiency. The high water splitting efficiency of Al:$SrTiO_3$[1] has invigorated research into oxide semiconductor doping mechanisms[2],[3]. Photoluminescence (PL) and transient absorption are frequently used for photocarrier lifetime and trapping studies, but it can be difficult to account for non-radiative recombination and surface effects. Cyclic voltammetry (CV) PEC measurements can be more effective for exploring the complexity of doping effects, since PEC measurements provide direct, quantitative estimates of the energy conversion efficiency.

Al-doping can be complicated because Al can substitute either a Sr or Ti atom in $SrTiO_3$. Depending on the doping site, the Fermi level will be pushed higher or lower. In the case of Ti-site doping, Al-doping can eliminate the Ti^{3+} recombination center[3] or create suitable in-gap states that reduces the photocarrier recombination rate[4]. The dominant doping site is affected by the oxygen activity and the doping level[5].

A doping level of <2% is sufficient for reducing the recombination rate without altering the band gap. However, such low doping levels are difficult to control in powder synthesis, along with other problems in studying powder photocatalysts such as defining the effective surface area and the distribution of particle size. Additionally, the intrinsic cation defect levels are also sensitive to synthesis conditions such as oxygen pressure and temperature[6]. Since the PEC efficiency is strongly dependent on the matching of the intrinsic defect level and the Al doping level, precise control of the synthesis conditions is required.

In this work, Al:$SrTiO_3$ thin films are grown by pulsed laser deposition(PLD), where the oxygen pressure can be accurately controlled over 8 orders of magnitude. The doping level of the thin film is controlled and assured by using PLD targets with different doping levels. This doping method can also be used to create a gradient doping level for systematic defect-dopant density balance studies. The main goal is to determine experimentally if the substitution site can be controlled by varying the growth condition. The chemical potential changes resulted from different growth conditions are observed by measuring the PEC water splitting activity by CV.

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