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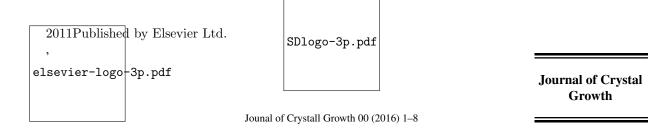
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#### **ACCEPTED MANUSCRIPT**



### Growth of Bi<sub>2</sub>Te<sub>3</sub> films and other phases of Bi-Te system by MOVPE

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#### Abstract

We have deposited films of Bi-Te system by atmospheric pressure MOVPE on (0001)  $Al_2O_3$  substrates with thin ZnTe or thick GaN buffer layers at different temperatures and Te/Bi ratio in the vapor phase. As-grown films were studied by X-ray diffractometry, SEM microscopy and Raman spectroscopy. To determine the elemental composition of the films, an energy dispersive spectrometer was used. Single-phase films of  $Bi_2Te_3$ ,  $Bi_4Te_5$ ,  $BiTe_5$ ,  $BiTe_6$ ,  $Bi_1OTe_9$ ,  $Bi_4Te_3$ ,  $Bi_3Te_2$  have been grown and growth parameter ranges for obtaining different phases were defined. It was found that under the same growth condition different phases of the Bi-Te system realize depending on the film's thickness. Thus, when growing of  $Bi_2Te_3$  films by MOCVD method the careful control of the phase composition is required.

*Keywords*: A1. Solid solutions, A1. X-ray diffraction, A3. Metalorganic vapor phase epitaxy, B1. Bismuth compounds, B2. Topological insulators

#### 1. Introduction

Recently thin films of bismuth telluride have been intensively investigated as a topological insulator (TI), a new material of condensed matter physics [1]. Moreover, versatile TI saturable absorbers, including those on the base of  $Bi_2Te_3$  nanoparticles, have been employed to passively mode-lock the fiber lasers at telecommunication wavelength regime [2, 3, 4]. For the latter application, thin films are needed and the high bulk resistance of TI is likely not required. Various deposition techniques, such as sputtering [5], thermal evaporation [6], electrodeposition [7], pulsed laser deposition [8], molecular beam epitaxy [9, 10] and metalorganic chemical vapor epitaxy [11, 12] have been developed to grow thin  $Bi_2Te_3$  films on different substrates.

Today to best of our knowledge there is no good quality phase diagram of the Bi-Te system, which would include different phases of the homologous series  $mBi_2 \cdot nBi_2Te_3$ , where m and n are numbers of  $Bi_2$  and  $Bi_2Te_3$  blocks per unit cell [13, 14, 15]. When depositing thin films of  $Bi_2Te_3$  other phases of the system Bi-Te may appear. The authors of the paper [16] have grown films with  $Bi_4Te_3$  phase in MBE system at the flows ratio of Te/Bi lower than 17. Caha et.al [17] observed the growth of BiTe phase on  $BaF_2$  substrates.

In this paper we report the observation of many transition phases from  $Bi_2Te_3$  to  $Bi_2$  when the growth temperature and the Te/Bi ratio in vapor phase are varied upon deposition of thin films in  $BiMe_3$ -Et<sub>2</sub>Te -H<sub>2</sub> system. We have found that the phases  $Bi_2Te_3$  (m=0, n=3,)  $Bi_4Te_5$  (m=1, n=5),  $Bi_{10}Te_9$  (m=6, n=9), BiTe (m=1, n=2),  $Bi_4Te_3$  (m=3, n=3),  $Bi_3Te_2$  (m=5, n=4) and  $Bi_2$  (m=3, n=0) of infinitive adaptive series  $mBi_2 \cdot nBi_2Te_3$  may be obtained. The formation of  $Bi_4Te_5$ , BiTe,  $Bi_4Te_3$  and  $Bi_3Te_2$  phases occurs over a wide range of Te/Bi ratio in the vapor phase at low temperatures. Thus, it is essential to identify and control the emergence of other phases of bismuth telluride when growing  $Bi_2Te_3$ . One should use with caution such a technological method as a low temperature deposition of films to suppress the free bonds of the substrate.

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