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Physics Procedia

Physics Procedia 81 (2016) 53 - 56

## 28th International Symposium on Superconductivity, ISS 2015, November 16-18, 2015, Tokyo, Japan

# Single-crystal growth and superconducting state of $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$

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#### Abstract

Single crystals of LaO<sub>0.5</sub>F<sub>0.5</sub>Bi(S<sub>0.8</sub>Se<sub>0.2</sub>)<sub>2</sub> were grown by a CsCl-flux method. Electrical resistivity  $\rho(T)$  measurements were performed to reveal its superconducting properties. The  $\rho(T)$  of the single crystal shows semiconducting-like behavior and superconducting transition at 3.4 K. The value is slightly higher than that of the polycrystalline sample with substitution amounts of x = 0.2 ( $T_c \sim 2.5$  K). From  $\rho(T)$  measurements in several magnetic fields,  $\mu_0 H_{c2}^{//ab}(0)$  and  $\mu_0 H_{c2}^{//c}(0)$  are estimated to be 17.2 T and 0.59 T, respectively. The superconducting anisotropic parameter  $\gamma$  is determined to be 29.2.

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Keywords: superconducto; BiS2-based compound

#### 1. Introduction

Since the discovery of superconductivity with superconducting transition temperature  $T_c$  at 8.6 K in the layered bismuth oxysulfide Bi<sub>4</sub>O<sub>4</sub>S<sub>3</sub> [1], other BiS<sub>2</sub>-based superconductivity have been studied. LaOBiS<sub>2</sub> crystallizes with a space group *P*4/nmm (No. 129) and this structure is composed of alternating superconducting BiS<sub>2</sub> layers and blocking LaO layers, which are similar to Fe-based superconductors. Substituting F for O induces superconductivity with  $T_c = 2.5$  K [2], as well as tetravalent elements (Ti, Zr, Hf and Th) for La [3]. In addition, by replacing S atoms

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to Se atoms,  $T_c$  is enhanced:  $T_c = 4.2$  K for LaO<sub>0.5</sub>F<sub>0.5</sub>BiSSe [4], 3.7 K for LaO<sub>0.5</sub>F<sub>0.5</sub>BiSe<sub>2</sub> [5]. The enhancement of  $T_c$  in Se doped compounds is induced by an in-plane chemical pressure [6].

In this article, we report the single crystal growth of  $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$  and its superconducting properties. In order to clarify superconducting state, we measured the electrical transport properties of  $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$  at low temperature and strong magnetic fields up to 11 T.

#### 2. Experimental Details

Single crystals of LaO<sub>0.5</sub>F<sub>0.5</sub>Bi(S<sub>0.8</sub>Se<sub>0.2</sub>)<sub>2</sub> were grown by a CsCl-flux method similar to the previous reports [5,6]. The starting materials of 0.8 g and CsCl of 5 g were mixed and sealed in an evacuated quartz tube. The tube was heated up to 880 °C in 10 h and to 900 °C in 2 h, then kept for 10 h and cooled down to 650 °C for 125 h. The obtained materials were washed by water, ethanol, and acetone in order to remove the flux materials. The obtained single crystals were plate-like shape with approximately  $1.0 \times 1.0 \text{ mm}^2$ . The crystal structure of the single crystal was examined by an X-ray diffraction (XRD) method using a conventional X-ray spectrometer equipped with Cu-K $\alpha$  radiation and a monochromator (RAD-2X, Rigaku). Electrical resistivity  $\rho(T)$  from 0.5 K to 300 K was measured under magnetic fields up to 11 T along the *ab*-plane and *c*-plane by dc-four-probe method in a <sup>3</sup>He cryostat (Oxford Heliox VL).

#### 3. Experimental Result

#### 3.1. X-ray diffraction

Figure 1 shows the XRD diffraction pattern of several single crystals  $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$  at room temperature. Only (00*l*) diffraction peaks were observed, indicating the crystallographic *c*-axis is perpendicular to the crystal plane. All the reflections can be indexed as the space group of *P*4/nmm and no extra peaks due to impurity phase can be detected from the XRD pattern. To estimate lattice constants, powder X-ray diffraction was performed by using powder crashed from single crystals. The lattice constants of  $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$  are obtained to be *a* = 4.0856 Å and *c* = 13.453 Å. These values are slightly larger than those of the single crystal  $LaO_{0.5}F_{0.5}BiS_2$ , which suggest that S atoms are substituted with Se atoms.



Fig. 1. XRD diffraction pattern of single crystals  $LaO_{0.5}F_{0.5}Bi(S_{0.8}Se_{0.2})_2$  at room temperature.

#### 3-2. Electrical resistivity

Figure 2 shows temperature dependence of the electrical resistivity  $\rho(T)$  of LaO<sub>0.5</sub>F<sub>0.5</sub>Bi(S<sub>0.8</sub>Se<sub>0.2</sub>)<sub>2</sub> down to 0.5 K with electrical current along the *ab*-plane. A semiconducting-like behavior was observed and superconducting transition was detected.  $T_c$  is defined as 50 % drop from residual resistivity  $\rho_0$  and determined to be 3.4 K.  $\rho_0$  is defined at just above  $T_c$  ( $\rho_0 = 3.5 \text{ m}\Omega \text{ cm}$ ). Compared with polycrystalline sample of LaO<sub>0.5</sub>F<sub>0.5</sub>Bi(S<sub>0.8</sub>Se<sub>0.2</sub>)<sub>2</sub> [7],

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