MLF Experimental Report	提出日 Date of Report
課題番号 Project No. 2012B0233 実験課題名 Title of experiment Possible shallow muonium state in barium titanate 実験責任者名 Name of principal investigator Takashi Ito 所属 Affiliation Advanced Science Research Center, Japan Atomic Energy Agency	装置責任者 Name of responsible person Prof. Yasuhiro Miyake 装置名 Name of Instrument/（BL No.） Muon D1 実施日 Date of Experiment 2012/12/10 9:00 – 2012/12/12 9:00 2012/12/13 9:00 – 2012/12/14 9:00

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）
Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

Barium titanate

$BaTiO_3$

Single crystal wafer ($20x20x0.5mm^3$)

2. 実験方法及び結果（実験がうまくいかなかった場合、その理由を記述してください。）
Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

The charge states of positive muons in $BaTiO_3$ were studied by the ZF, LF and TF-μSR techniques using a single-pulsed muon beam. Typical ZF-μSR spectra at cryogenic temperatures are shown in Fig. 1. Below 80K, a fast relaxation component emerges and its fraction gradually increases with decreasing temperature. This cannot be accounted for solely by the magnetic dipolar interactions with adjacent nuclei, thereby ascribed to a muon accompanied by a localized unpaired electron around the muon site. This is naively interpreted as an electron-muon bound state, namely, a muonium Mu^0. A muon-induced polaronic center, proposed by Cox *et al.* in transition-metal oxides, is also possible [1]. Hereafter, this defect center associated with an implanted muon is referred to as Mu^0/P.

Figure 2 shows the temperature dependences of the Mu^0/P fraction (open circle) and the diamagnetic μ^+ fraction (closed circle) obtained by fitting the ZF-μ^+SR spectra to a sum of two exponential functions. The TF-μ^+SR spectra were also analyzed with a sum of two TF relaxation functions with exponential envelopes. The Mu^0/P and μ^+ fractions in TF, plotted on Fig. 2 with open and closed squares, approximately agree with those in ZF. As the temperature increases, the diamagnetic fraction grows at the expense of the Mu^0/P fraction. This indicates

2. 実験方法及び結果（つづき） Experimental method and results (continued)

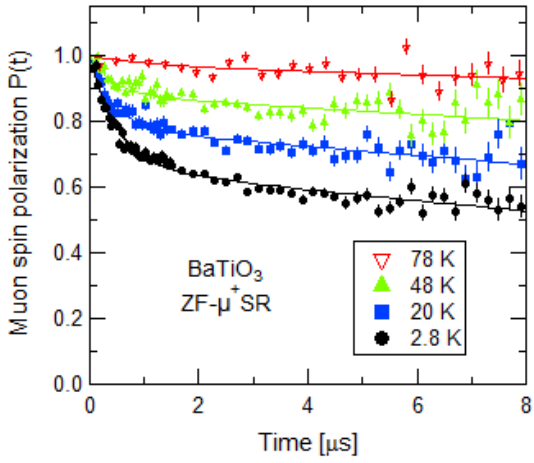

Fig. 1

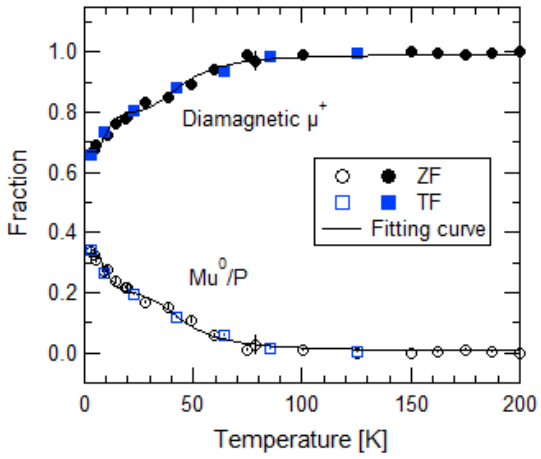

Fig. 2

the thermal excitation of a weakly bound electron into the conduction band at the moderate temperatures and implies that the Mu^0/P center forms shallow donor levels.

To determine the effective donor depth, the temperature dependences of the Mu^0/P and diamagnetic fractions were analyzed using an ionization model [1]. A reasonable fit, shown in Fig. 2 by solid curves, was obtained under the assumption of the existence of two different defect centers, as in the case of ZnO [2]. The ionization energies E_i of the defect centers were estimated to be 4.3(6) meV and 19(3) meV, respectively, which are the same order of magnitude as those of shallow-donor muonium in a series of binary semiconductors and insulators. The effective donor depth E_d is empirically estimated to be in the range $E_i < E_d < 2E_i$. The shallow effective donor depth suggests that the weakly bound electron at cryogenic temperatures is excited into the conduction band at room temperature and behaves as a free carrier. Therefore, the implanted positive muon, by analogy a hydrogen impurity, will cause unintentional *n*-type conductivity in $BaTiO_3$.

[1] S. F. J. Cox *et al.*, J. Phys.: Condens. Matter **18**, 1079-1119 (2006).

[2] K. Shimomura, K. Nishiyama, and R. Kadono, Phys. Rev. Lett. **89**, 255505 (2002).