（ ※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。 ）

MLF Experimental Report

提出日 Date of Report	
課題番号 Project No. 2012A0063	装置責任者 Name of responsible person Prof. Yasuhiro Miyake
実験課題名 Title of experiment μ^+SR and μ^-SR studies of hydrogen behavior in a new oxyhydride $BaTi(O,H)_3$	装置名 Name of Instrument/（BL No.） Muon D1
実験責任者名 Name of principal investigator Takashi Ito	実施日 Date of Experiment 2012/05/20-2012/05/25
所属 Affiliation Japan Atomic Energy Agency	

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.

(1) $BaTiO_{2.5}H_{0.5}$, 3g, a pellet wrapped with an Al foil

(2) $BaTiO_3$, 1.2g, two pieces of single crystalline samples wrapped with Al foils

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.

(1) μ^+SR in $BaTiO_{2.5}H_{0.5}$

A ϕ24 mm pellet of $BaTiO_{2.5}H_{0.5}$ was mounted on a silver sample holder of a conventional ^{4}He flow cryostat. ZF/LF-μ^+SR measurements were performed at the D1 area using a double-pulsed muon beam. Figure 1 shows a typical ZF-μ^+SR spectrum at low temperatures below 100 K. The shaded area indicates an instrumental background determined by using a reference sample. Almost all the remaining signal damps significantly within the time window, indicating that *hydride ions randomly distribute over the whole volume of the specimen*. An oscillating pattern was clearly observed, suggesting formation of a ^{1}H-μ spin entangled state [R. Kadono *et al.*, PRL **100**, 026401 (2008)]. The ZF-μ^+SR spectra at low temperatures were successfully fitted with the following function, $A_s[p_{2S}\, e^{-\lambda t}\, G_{2S}(t;\omega_d)+(1-p_{2S})\exp(-\Delta^2 t^2)] + A_{BG}$, where A_s and A_{BG} are partial asymmetries for the sample and the instrumental background, p_{2S} is the fractional weight for the two spin entangled state, $G_{2S}(t)$ is the relaxation function for the two spin entangled state, ω_d is the angular frequency, and λ and Δ are the relaxation rates. The best fit for the data at 30 K is shown in Fig. 1 with the solid line. The fitting parameters are almost constant below 100 K. An H^--μ distance of the two spin entangled state was estimated to be ~0.17 nm from ω_d.

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

This agrees well with a distance between an interstitial protonic impurity site and a second nearest neighbor anion site in pure $BaTiO_3$. Thus, we conclude that *muons implanted into* $BaTiO_{2.5}H_{0.5}$ *stay at the interstitial site as* μ^+ *as shown in Fig. 2 and microscopically coexist with the* H^- *ions at the anion sites in the same lattice for at least several tens of* μs. This suggests a possibility to form *hydroxyhydrides*, where both H^+ and H^- are present in the same lattice.

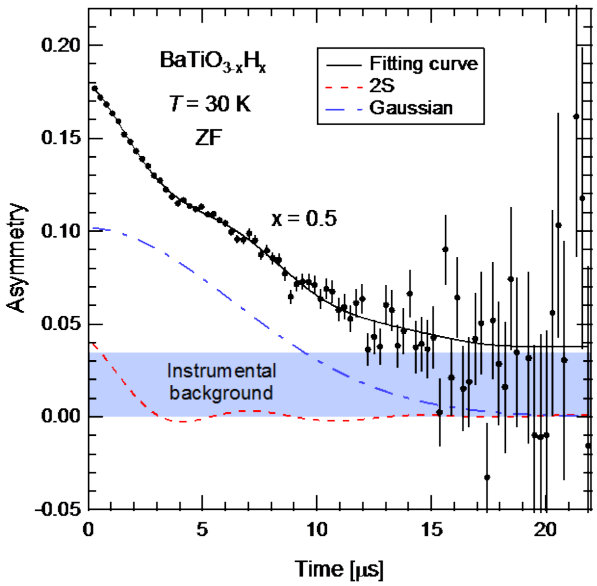

Fig.1: The ZF-μ^+SR spectrum of $BaTiO_{2.5}H_{0.5}$ at 30 K.

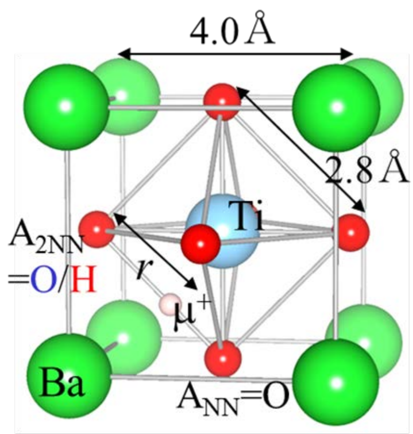

Fig.2: A schematic illustration of the μ^+ site in $BaTiO_{2.5}H_{0.5}$ below 100 K.

(2) μ^+SR in $BaTiO_3$

ZF/LF/TF-μ^+SR measurements on pure $BaTiO_3$ were performed at the D1 area in order to compare with the μ^+SR results in $BaTiO_{2.5}H_{0.5}$. Two pieces of single-crystalline samples of $BaTiO_3$ covering the cross-sectional area of 10x20mm^2 was mounted on a silver sample holder of a conventional ^{4}He flow cryostat. Fast muon spin relaxation was observed at low temperatures, indicating muonium formation. A LF-decoupling study revealed that the hyperfine coupling constant of the muonium in $BaTiO_3$ is considerably smaller than that in vacuum, suggesting a shallow muonium state. We also carried out TF-μ^+SR measurements in a magnetic field of 123 Oe to obtain spectroscopic evidence of the shallow muonium state. A frequency splitting suggesting the shallow muonium state was observed, but this is not so clear due to the double pulse structure of the muon beam.

*A new proposal for shallow muonium hunting in pure $BaTiO_3$ was submitted to J-PARC PAC in order to obtain clear evidence of the shallow donor state in $BaTiO_3$. The proposal was recently approved.

(3) μ^-SR

The μ^-SR measurements planned in the proposal has not been done yet since the allocated beamtime was not sufficient for such an experiment. An up-grade of the spectrometer or beam intensity is necessary to perform μ^-SR measurements in a few days of beamtime.