MLF Experimental Report	提出日 Date of Report 2012/03/05
課題番号 Project No. 2010A0059 実験課題名 Title of experiment MuSR study on isolated hydrogen charge state in oxgen deficient SrTiO3 実験責任者名 Name of principal investigator Koichiro Shimomura 所属 Affiliation KEK-IMSS	装置責任者 Name of responsible person Yasuhiro Miyake 装置名 Name of Instrument/(BL No.) D1 実施日 Date of Experiment 2010/5/25,26,28

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.

$SrTiO_3$

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.

Hydrogen is a ubiquitous impurity in most semiconductors, including elemental (e.g., Si), compound (e.g., GaAs) one. In these systems, hydrogen is known to be amphoteric, forming an acceptor level in *n*-type and a donor level in *p*-type materials. In contrast, hydrogen can lead to electron conduction in some wide gap semiconductors such as ZnO. Motivated by this theoretical prediction, μSR Studies on ZnO and electron-nuclear resonance measurements on *n*-type ZnO supported the hypothesis that hydrogen is a shallow donor. Several other studies show evidence that hydrogen is related to *n*-type conductivity in ZnO.
These observations raise the question of what is the basic systematic at work here: if H can be incorporated into some materials, which one will be doped by H (i.e., become conductive) and which will not? Experimentally, μSR studies have played pioneering roles on this subject. While the dynamical aspects (e.g., diffusion) may be considerably different between Mu and H due to the light mass of Mu (*mass*~ $1/9m_p$), the local electronic structure of Mu is virtually equivalent to that of H after a small correction due to the difference in the reduced mass (~0.4%). Recently, a novel Mu state having an extremely small hyperfine parameter (10^{-4} times smaller than the muonium in vacuum) and a low ionization energy (~10meV) was reported in several compound semiconductors including CdS , ZnSe, InN and GaN, implying that Mu (and hence H) can act as a donor in these materials. On the other hand, considerable theoretical studies were also performed. Very recently Iwazaki and Tsuneyuki performed interesting theoretical work on $SrTiO_3$ related these subjects.

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

The physical properties of $SrTiO_3$ have been extensively investigated over the years mainly due to its potentiality for device applications, especially, in ceramic condensers, photo catalysis, and sensors. Electrically conducting $SrTiO_3$ crystals are often obtained by annealing in highly reducing, hydrogen containing atmospheres. The role of hydrogen is complicated. Subsequent anneals at 800 °C in forming gas, which contained 10% hydrogen, returned the crystals into the insulating, Understanding the roles of hydrogen related these phenomena in $SrTiO_3$ is also of technical importance. Forming gas, which contains hydrogen, is used in semiconductor device processing and causes a drastical change in the leakage current of $SrTiO_3$ films

However, some theoretical work suggests interstitial hydrogen has been suggested to act as a shallow donor in $SrTiO_3$ and related materials. Thus both oxygen vacancies and hydrogen may potentially contribute to the electrical conductivity of $SrTiO_3$. Iwezaki and Tsuneyuki theoretically point out the hydrogen in oxygen deficient $SrTiO_3$ can form the negative charge state, therefore, it might suppress the electrical conductivity (passivation). This theoretical picture well explains the phenomena mentioned above.

To check these theoretical predictions by experimentally, we performed the MuSR measurements. For the first stage the comparison of the muon spin relaxation rate in TF and ZF field between normal and annealed $SrTiO_3$ samples. As shown in Fig.1, there is a drastic change for typical time differential spectrum between them at the room temperature. However, in that time, beam time and intensity were very limited, therefore, we could not performed detailed temperature dependence.

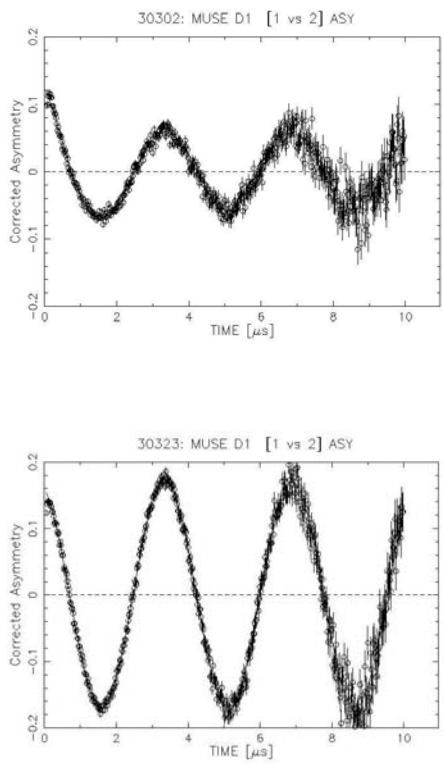

Fig.1 TF spectrum of Oxygen defficient (above) and normal $SrTiO_3$ at 300K.