

 MLF Experimental Report	提出日 Date of Report 2012/03/05
課題番号 Project No. 2010B0023 実験課題名 Title of experiment MuSR study on isolated hydrogen charge state in oxygen deficient SrTiO ₃ 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 2011/2/9,11,12

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.
<p>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 <i>n</i>-type and a donor level in <i>p</i>-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 <i>n</i>-type ZnO supported the hypothesis that hydrogen is a shallow donor. Several other studies show evidence that hydrogen is related to <i>n</i>-type conductivity in ZnO.</p> <p>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 ($mass \sim 1/9m_p$), 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 ($\sim 0.4\%$). 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 ($\sim 10\text{meV}$) 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 SrTiO₃ related these subjects.</p>

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 $800\text{ }^\circ\text{C}$ 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).

In this year we performed temperature dependence of the relaxation rate in oxygen deficient and normal STO. As shown in Fig.1, there is a drastic change of the relaxation rate was observed in oxygen deficient STO. However, no significant change was observed in normal STO. These phenomena strongly indicate the spin and/or charge exchange process of negatively charged muonium occurs above 180K. The high transverse field measurement will help to clarify these phenomena.

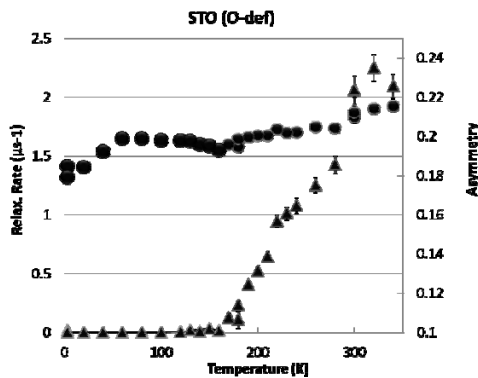


Fig.1 Temperature dependence of asymmetry (closed circle) and relaxation rate (closed triangle) of oxygen deficient SrTiO_3 at TF(20G) measurements..