3-4 Hydrogen-Induced Insulation Degradation of Ceramic Capacitors

— Microscopic Evidence Provided by Muons —

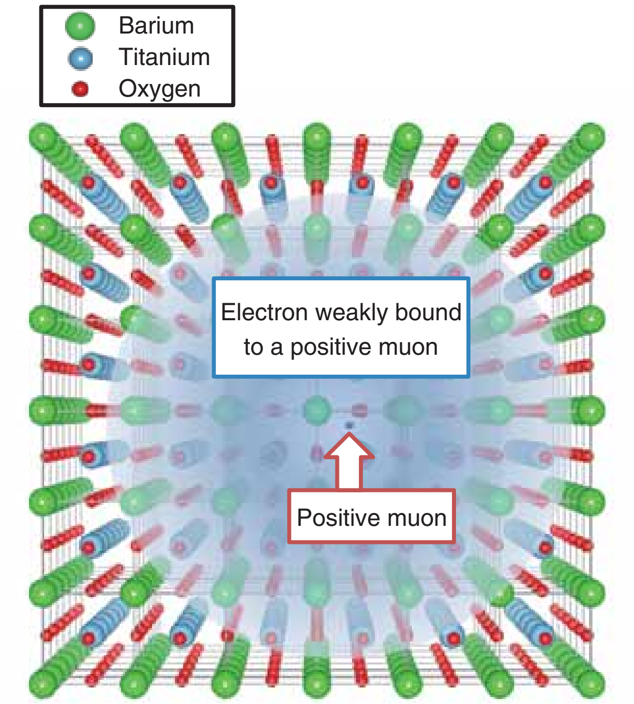

Fig.3-8 A schematic of the muon-electron bound state in $BaTiO_3$
The widely spread electron orbital suggests that the electron is very weakly bound to the positive muon in $BaTiO_3$.

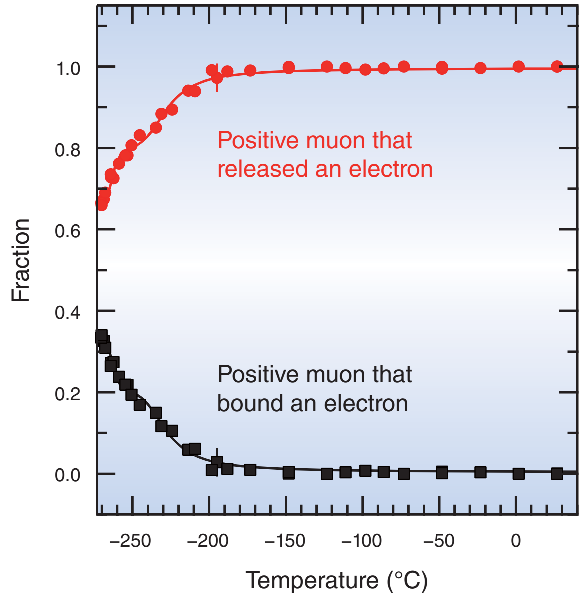

Fig.3-9 Electron release from the hydrogen-like muon-electron bound state in $BaTiO_3$
The black squares and red circles represent the fractions of positive muons with and without a bound electron, respectively. The solid curves are the best fits to an ionization model.

Multilayer ceramic capacitors (MLCCs) are fundamental components of electronic circuits and are indispensable for state-of-the-art electronic devices. Since the performance of MLCCs heavily depends on that of dielectric materials, controlling the quality of dielectric materials is vitally important. As the internal electrodes of typical MLCCs contain an easily-oxidizable element, the annealing process to unify the dielectrics and electrodes is performed in a reducing atmosphere containing hydrogen (H). On the other hand, this process can cause insulation degradation, depending on the annealing conditions. Intensive studies are under way to identify the origin of this degradation.

In this work, we focused on the risk of H incorporation into dielectrics in the annealing process, and studied the behavior of H in a typical dielectric material, barium titanate ($BaTiO_3$). The electronic structure of H impurities in $BaTiO_3$ is not obvious, since H can assume various charge states in materials. To tackle this issue, we used positive muons in place of H. It is well established that the electronic structure of a muon-electron bound state is identical to that of H except for small isotope corrections. Thus, we can imitate H impurities in $BaTiO_3$ with positive muons and selectively probe their influences. Another advantage of using a muon is its high sensitivity; one can obtain microscopic information corresponding to H impurities in the dilute limit, which is difficult to access by other experimental techniques.

A positive muon beam was irradiated to a $BaTiO_3$ single crystal at the J-PARC muon facility, and the local electronic structure around the implanted muons was investigated by means of the muon spin rotation technique. We observed a signal from muons that weakly bound an electron below −190 °C, which suggested that the electron orbital was widely spread, as shown in Fig.3-8. The weakly bound electrons were gradually released with increasing temperature, as shown in Fig.3-9. The released electrons could move freely around the crystal and led to electric conductivity, thus decreasing the insulating performance of $BaTiO_3$. Hydrogen impurities in $BaTiO_3$ are also thought to release electrons according to a similar mechanism, resulting in insulation degradation at device operating temperatures.

Reference

Ito, T. U. et al., Shallow Donor Level Associated with Hydrogen Impurities in Undoped $BaTiO_3$, Applied Physics Letters, vol.103, issue 4, 2013, p.042905-1-042905-4.