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Search for a space charge layer in thin film battery materials with low-energy muons

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In an all solid state Li-ion battery, it is crucial to reduce ionic resistivity at the interface between the electrode and the electrolyte in order to enhance Li+ mobility across the interface, because Li⁺ ions naturally drift across such interface. In particular, recent first principles calculations predict the presence of a space-charge layer (SCL) at the interface because of the difference in the Li⁺ chemical potential between the two materials [1], as in the case for the interface between a metal and a semiconductor in electronic devices. However, the presence of SCL has never been experimentally observed. We have therefore initiated series of studies for direct observation of SCL in different cathode battery materials, exploiting the unique depth-resolved features of the LEM- μ ⁺SR experimental technique.

Our first attempt in a fresh multilayer sample, $Cu(10 \text{ nm})/Li_3PO_4(50 \text{ nm})/LiCoO_2(100 \text{ nm})$ on a sapphire substrate, revealed a gradual change in the nuclear magnetic field distribution width, originating from the nuclear fields, as a function of implantation depth even across the interface between Li_3PO_4 and $LiCoO_2$. This implies that the change in the field distribution width at SCL of the sample is too small to be detected by LEM- μ^+ SR. Since the SCL is expected to be amplified by charge-discharge reactions, future attempts to observe the space-charge layer will be conducted on multilayer samples after charge and discharge.

[1] J. Haruyama et al., Chem. Mater. 26, 4248 (2014).

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