Toward Operando Observation of Li-Ion Battery
by Scanning Transmission Electron Microscopy

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All-solid-state Li-ion battery (LIB) is a promising device because of its high energy density, high safety, and long lifetime. For improvement of the battery performance, one of the key issues is to clarify the diffusion mechanism of lithium (Li) ions in cathode materials, such as $LiCoO_2$. Previous studies suggested that the Li-ion diffusion is influenced by the local structures, such as grain boundaries and defects. However, the influence has not been clarified experimentally yet. Transmission electron microscopy (TEM) is a powerful way to identify nanometer- to atomic-scale structures, but has a problem of sample damage due to the electron beam irradiation.

Here, we focused on the moiré method of scanning TEM (STEM). It can detect a slight change (~0.1%) of strain distribution with low electron beam irradiation (~1/1000 of the conventional S/TEM images). Meanwhile, as for $LiCoO_2$, the *c*-plane distance changes dependent on the Li amount. The Li-ion diffusion in $LiCoO_2$ can be estimated by the strain distribution change during the battery charging.

We observed a $LiCoO_2$ thin film epitaxially grown on a $SrTiO_3$(111) substrate. Fig. 1 is an atomic resolution STEM image of $LiCoO_2$ viewed along [010] direction. As shown in bright peaks in Fig. 1, the cobalt (Co) atoms are aligned horizontally, forming the (003) planes. A STEM moiré image is shown in Fig. 2. A pattern with an interval of ~10 nm can be seen. The pattern is a moiré caused by the interference between the scanning grating spacing (502 pm) and the (003) lattice spacing (472 pm). The grain boundaries can be distinguished from the contrast of the moiré image.

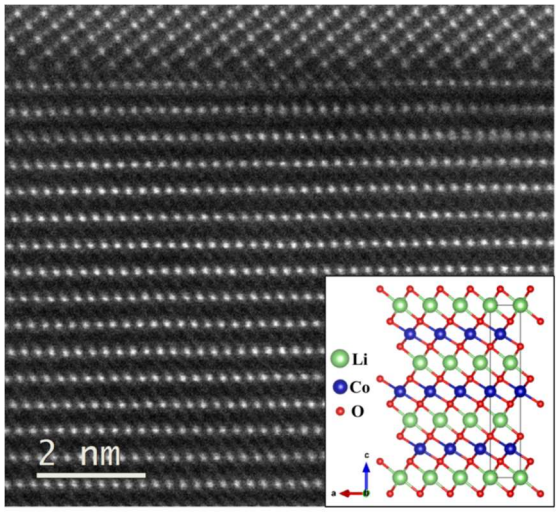

Fig. 1: STEM image of $LiCoO_2$

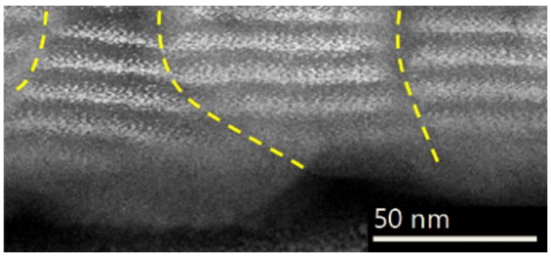

Fig. 2: STEM moiré pattern (dashed lines indicate the grain boundaries)

A micro battery sample was prepared and contacted with the electrodes of the device. The electrochemical properties of the micro battery were examined. However, the results showed a short circuit. The cause seems to be the tungsten deposition used in the sample processing. By overcoming the problem, we will be able to establish the method of the *operand* STEM moiré observation of Li-ion diffusion.

Keywords: Li-ion battery, ion diffusion, *operand* STEM, STEM moiré