Bragg Coherent Diffractive Imaging of non-equilibrium strain dynamics in lithium-excess battery nanoparticles

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Performance of commercially used battery materials is one of the limiting factors in many modern technologies from vast variety of portable electronics to drones and spacecrafts[1]. It is commonly accepted, that the functional characteristics of the battery material, such as capacity and lifespan are determined by the structure and chemistry at a single particles level[2]. Currently, Bragg Coherent X-ray Diffractive Imaging (BCDI) is the only available technique that allows probing the structural dynamics inside individual nanoparticles during battery operation[3-4].

In this work, we present results of in-situ BCDI experiment with lithium-rich layered oxide (LRLO), one of the leading candidates for the next-generation energy storage cathode material. Despite a high interest from the scientific community, the understanding of the mechanisms underpinning oxygen-redox reactions and voltage fade in LRLO remain incomplete and prevents commercialization. Recent studies suggested a link between anomalous oxygen activity in LRLO and significantly higher rate of dislocation formation[5]. The aim of our study was to understand which of these process drives the other.

We performed in-situ Bragg CDI measurements of LRLO coin cell batteries for C/10 and C/5 charge rates. Comparing the rate of the lithium extraction with the speed of ionic diffusion we argue that C/10 charge rate corresponds to steady dynamics, while for C/5 the dynamics is non-equilibrium. Fig.1 shows Bragg CDI reconstruction for C/5 measurement at 4.1 V (a), the relative increase of the partial elastic energy (b) and the dislocation density (c). The remarkable 3-fold symmetry of the deformation field, which is specific to LRLO material, fades away during the charge and comes back after the discharge. Our results allow distinguishing if the dynamics of dislocation network is predominantly driven by the local variations of the chemical potential or the elastic energy landscape. We also suggest that the speed of dislocations is limited by the speed of lithium diffusion.

![Bragg CDI reconstruction](image)

**Figure 1.** Bragg CDI reconstruction(a), elastic energy density(b), and dislocation density (c) for LRLO.

**References**