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Determination of Crystallinity in $Li_{1-x}Mg_xMn_2O_4$ Nanocrystals Based on Diffraction Patterns Correlation Analysis and Strain Mapping

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Crystallinity refers to the degree of crystal order. In some materials such as polymers, the order and disordered regions are clearly differentiated by the alignment of molecules, and determination of crystallinity can be simply performed by diffraction analysis. However, crystallinity in many important nanocrystalline materials are much harder to determine using traditional approach. Here, we describe a scanning electron nanodiffarction (SEND) method [1-4] for the determination of crystallinity in nanocrystals. The method allows a determination of strain distribution in nanocrystals, as well as local orientation and phase changes, which we demonstrate for $Li_{1-x}Mg_xMn_2O_4$ nanocrystals, a potential Mg battery material**.**

The SEND experiments were carried out using a Themis Z STEM (Thermo Fisher Scientific) operated at 300kV. Spot-like diffraction patterns were recorded using an electron probe of a semi-convergence angle of 0.46 mrad and a probe size of 1.7 nm in FWHM (full-width at half-maximum) and a CMOS camera (Ceta Camera, Thermo-Fisher). The diffraction patterns (DPs) described in Figure 1 were acquired using a 45x50 pixel scan over a sample area of 90x100 nm^2. The total acquisition time is ~30 mins and during this period the sample drift is automatically corrected using the TIA software. The SEND patterns were first analyzed for their similarities by the pattern matching method and for strain. In diffraction pattern correlation analysis, a correlation map is created (Figure 1c), where the distribution of similar diffraction patterns is identified. The similarity of DPs is quantified by the value of normalized cross correlation coefficient (NCC) calculated for two individual diffraction patterns. The value of an NCC calculation between two DPs will range from -1 to 1 with NCC = 0 indicating complete dissimilarity and NCC = 1 indicating complete similarity. A threshold value of NCC, or 'correlation threshold' (CT), is defined. Two DPs are similar if their NCC is greater than CT. A group is defined next, which collects all similar pattern. The DPs in a group is then averaged to obtain the group average pattern (GP). Next, the experimental DPs are compared to all GPs, and sorted into a group by the highest NCC. This process can be repeated to improving the grouping and sorting process.

For strain analysis, the positions of strong beams, which include the center beam, were determined using Hough transform method for detecting disk position at subpixel precision [5]. The method works by applying first the Sobel filter to the DP to filter out the disk edge. Then, circular Hough transform is applied the filtered pattern, which transform the filtered pattern of edge circles into a pattern of Hough transformation peaks, with each peak marking the position of a detected circle. The peak position was measured by fitting using a Lorentzian peak model. This measurement was applied to all 9 selected beams, and a 2D reciprocal lattice was then determined from the measured disk positions. Strain for a given lattice plane was obtained using the calibrated camera constant and reference lattice. The above analysis strategy will be discussed and the information obtained will be compared based on the analysis of $Li_{1-x}Mg_xMn_2O_4$ nanocrystals. Specifically, we will discuss the use of electron diffraction patterns for the determination of Mg inserted into the nanocrystal [6].

References:
[1] Zuo, J.M. and J.C.H. Spence, "Advanced Transmission Electron Microscopy: Imaging and Diffraction in Nanoscience." 2016: Springer.
[2] Tao, J. et al., Physical Review Letters **103** (2009), p. 097202.
[3] Kim, K.H. et al., Micron **71** (2015), p. 39.
[4] Meng, Y. and Zuo, J.M., ICUrJ **3** (2016), p. 300.
[5] R. Yuan, Y. Meng, J. Zhang, J.-M. Zuo, Microscopy and Microanalysis **23** (2017), p. 180
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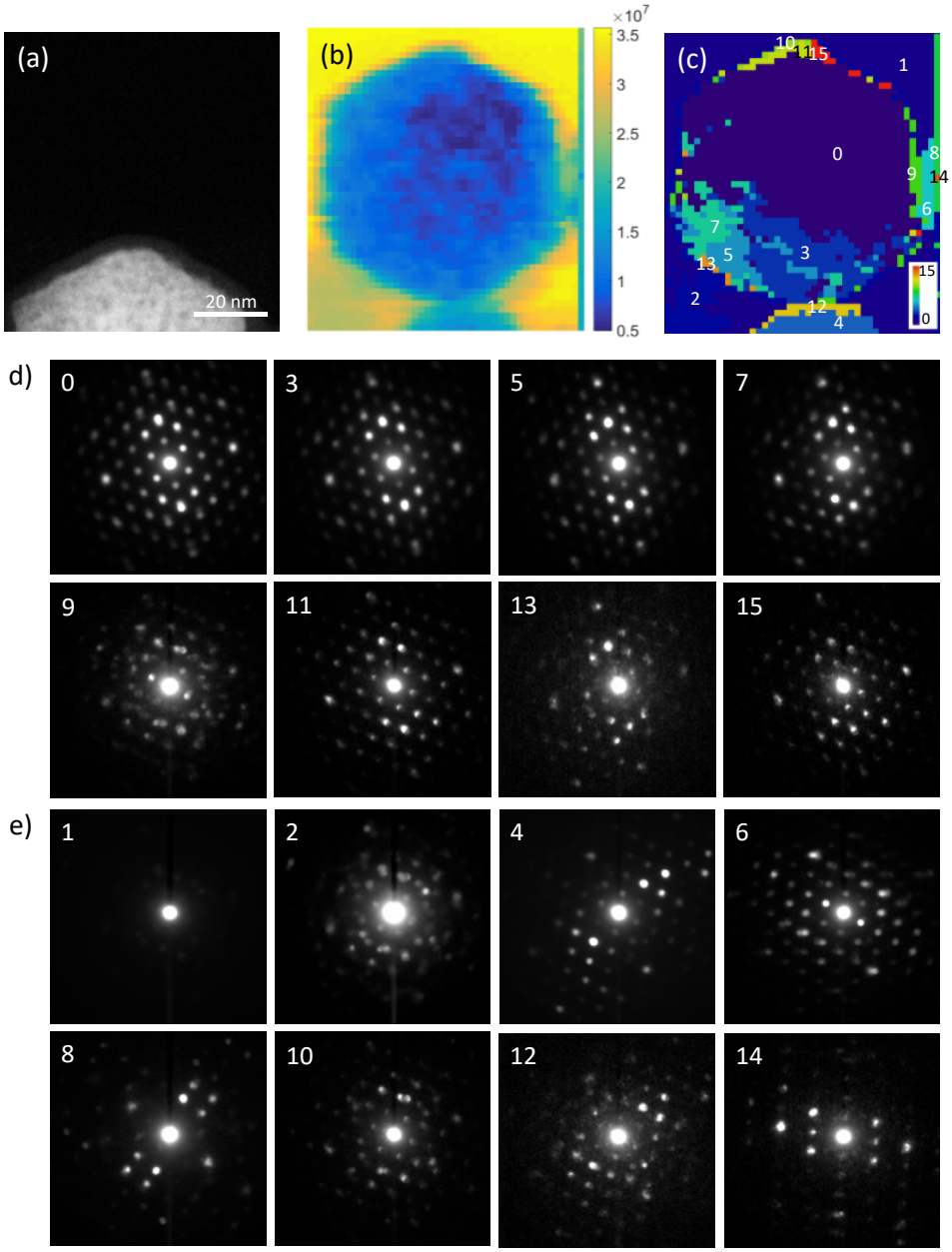

Figure 1. (a) STEM image of $Li_{1-x}Mg_xMn_2O_4$ where SEND was collected, (b) Virtual bright field image from SEND. (c) Correlation image. (d) Typical diffraction patterns from regions inside the center nanocrystal as marked in c). (e) Other DPs. CT=0.5 was used here.