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Cepstral Scanning Transmission Electron Microscopy Imaging of Disordered Crystals using Coherent Diffuse Scattering

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The development of four-dimensional (4D) scanning transmission electron microscopy (STEM) using fast detectors has opened-up new avenues for imaging defects, which ranges from being relatively simple, like virtual bright- or dark-field imaging, to being sophisticated, such as strain mapping using Bragg diffraction or Kikuchi/HOLZ lines. Here we introduce a new 4D-STEM technique, called Cepstral STEM, for imaging defects in crystals using electron diffuse scattering. In contrast to analysis based on Bragg diffraction, which measures the locally averaged d-spacings, elastic electron diffuse scattering can detect disruptions to the lattice. The disruptions, which gives rise to different types of diffuse scattering, include short range order, dislocations, stacking faults, and lattice distortion from crystal disorder. With the latter defines the atomic displacements from the average lattice sites, which differs from lattice strain that gives the change in local lattice parameters.

The challenge in using electron diffuse scattering for imaging comes from their relatively weak intensity and the inevitable inelastic scattering background. To solve the inelastic scattering problem, we use coherent electron nanodiffraction where interference gives the laser speckle like features in the recorded diffraction patterns. The inelastic background, which is incoherent, is filtered out by coherent scattering [1]. To overcome the weak signal issue, we employ pixelated detectors with large dynamic range to capture electron diffuse scattering

The harmonic signals in coherent electron diffuse scattering are detected through the difference Cepstral analysis method [2, 3], where we subtract the Cepstrum of a region averaged nanodiffraction pattern from that of a local pattern. We show theoretically that this subtraction removes Bragg diffraction from the diffraction pattern and provide Patterson function analysis of diffuse scattering. We demonstrate our method on dislocation defects and severely distorted high entropy alloys. The results are compared with strain maps calculated with from Bragg peak position in the same set of diffraction patterns. We demonstrate that strain maps and Cepstral STEM provide complementary information about deformed and distorted crystals at nm spatial resolution.

Figure 1 is an example of application to the high entropy alloy, $Al_{0.1}CoCrFeNi$. HEAs are comprised of at least five elements in an equal or near-equal atomic fraction, and the mismatch in the atomic radii of different elements results in a large number of atoms being displaced from their ideal positions (lattice distortion). Consequently, severe lattice distortion has been suggested as one of the four core effects in HEAs [4, 5]. However, the complex chemical and lattice disorder in the HEAs present a major challenge for their characterization [4]. In this presentation, we will demonstrate how Cepstral STEM and strain mapping together provide a full picture of distorted lattice in $Al_{0.1}CoCrFeNi$.

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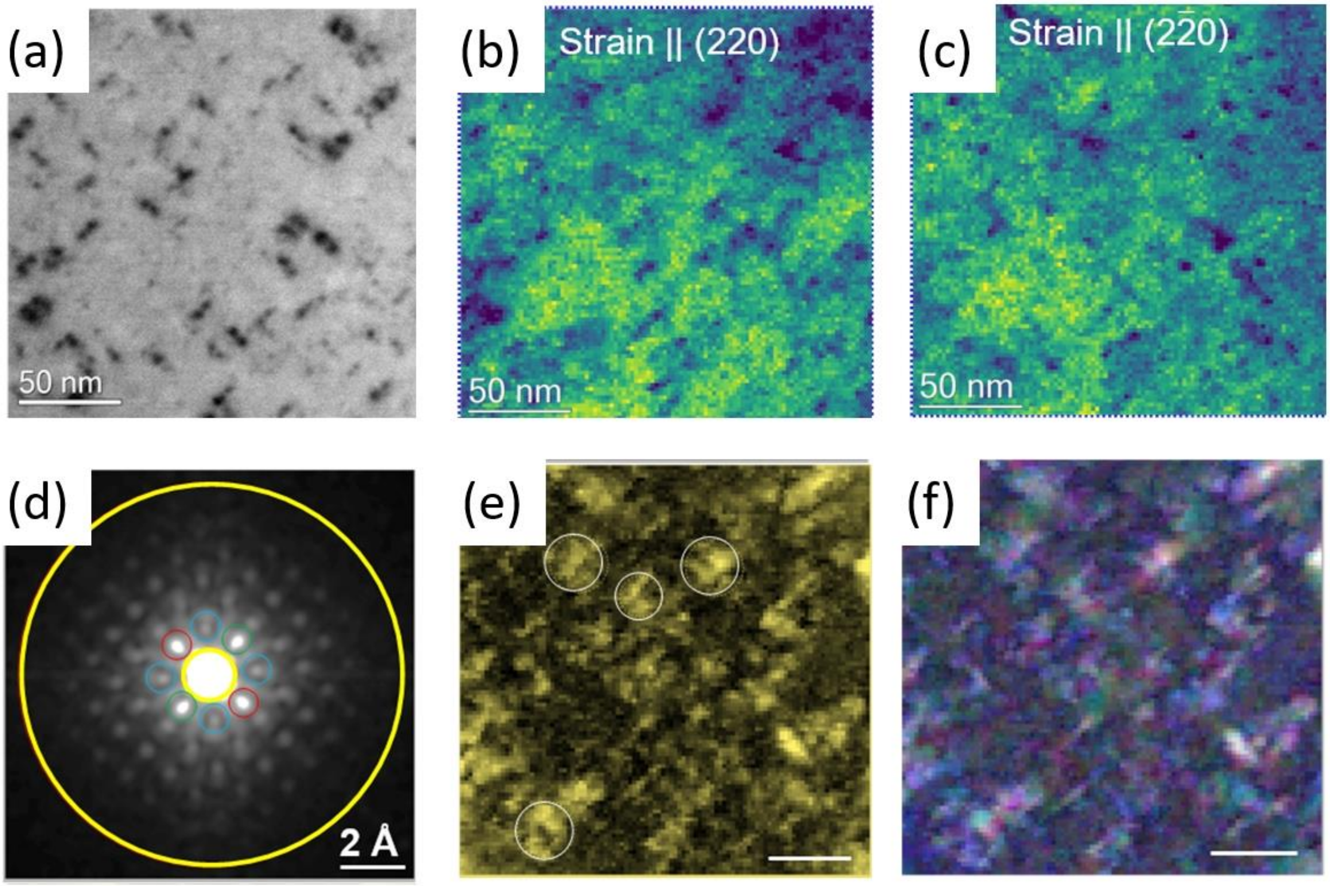

Figure 1. Fig. 1 (a) HAADF-STEM image of an area in Al0.1CoCrFeNi where 4D-STEM was performed using a 100x100 scan and a 2 nm step size. (b) and (c) Strain maps constructed from measuring the displacement of Bragg reflections in the 4D-STEM dataset. (d) Area averaged Cepstrum. (b) Cepstral ADF image obtained by integration between the marked yellow circles in (d). (f) Composite of Cepstral dark-field images obtained by integrating intensities within red, green and blue colored circles, for R, G and B colors. Scale bars in (e and f), 20 nm.

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