Direct observation of charge modulation in nanoprecipitates by 4D STEM

Dr. Juhyun Oh[1], Prof. Young-Woon Kim[1], Prof. Cheol-Woong Yang[2], Prof. Cheol-Hee Ahn[1], Prof Miyoung Kim[1]
[1]Department of Materials Science and Engineering, Seoul National University, Seoul, Korea, [2]Department of Materials Science and Engineering, Sungkyunkwan University, Suwon, Korea

Background incl. aims
Charge modulations induced by structural, compositional and electronic variation in a solid can be investigated using various analytical techniques, including scanning tunnelling microscopy/spectroscopy, Raman spectroscopy, and recently available four-dimensional scanning transmission electron microscopy (4D-STEM), depending on the nature of the modulation. We explore the interplay between charge density modulation and structural distortion in nanoscale moiré structures, $MgZn_2$ nanoprecipitates imbedded in Al matrix using 4D-STEM.

Methods
A series of diffraction patterns by 4D-STEM offers a wide range of information, which includes local electric fields and charge density, in addition to atomic configurations. We integrated intensities at different scattering angles and directions to extract various types of images. Template matching was employed to the real space, depth-sectioned STEM images to locate the accurate atomic positions, which facilitated the mapping of local strain distributions within the imbedded nanostructures. Density functional theory (DFT) calculations are performed to find the origin of the charge modulation and multislice simulations are further employed to compare the experimental results with the theoretical prediction.

Results
We present center of mass (COM) images derived from the in-plane diffraction patterns of the MgZn2/Al heterostructure. Interestingly, the images clearly reveal charge density modulations which are highly dependent on the depth of the probe position in the specimen. Moreover, strain mapping within the Al matrix is intricately linked to charge modulation, suggesting the potential influence of structural distortion on the charge modulation. The periodicity and the location of the modulation are further analyzed to determine the correlation with the moiré supercell structures.

Conclusions

A series of multislice simulations generated numerous COM images, encompassing single atoms to stacks of multi-layers, at different probe depths. We propose that the primary contribution to the modulated charge distribution originates predominantly from the interface area between the $MgZn_2$/Al heterostructure. Furthermore, we demonstrate how manipulation of COM images provides more detailed insights into the structural and compositional variations within materials.

Keywords:

4D-STEM, Charge, Nanoprecipitates