

## Dynamical Effects on Atomic-Resolution Imaging and Diffraction of the Tetragonal and Orthorhombic Phases of LaFeAsO

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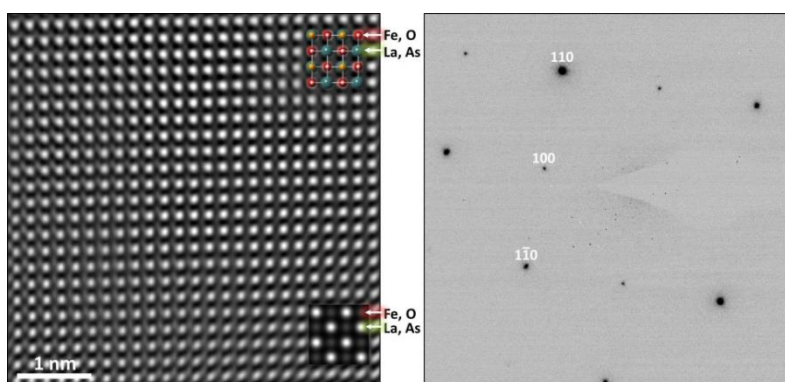
The discovery of high- $T_c$  superconductivity in the lightly-doped iron pnictides has led to an explosion in theoretical, synthetic, and characterization work [1,2]. Doping in these materials suppresses the temperatures at which the crystallographic phase transition and antiferromagnetic (AF) ordering occur relative to the undoped parent compounds. At sufficient doping levels, emergence of the superconducting dome is observed with the suppressed AF and so-called nematic states bordering this transition [3]. Of particular interest are the *1111*-type compounds, which have been found to exhibit  $T_c > 50$  K. Consequently, efforts have focused on elucidating the origin of such emergent properties in these and other strongly-electron correlated materials. Results suggest that there is a complex interplay between structural transformations and spin/charge/orbital ordering, the precise nature of which remains unknown with respect to the superconducting state [4].

With the advent of  $C_s$ -corrected microscopy along with advances in *in situ* TEM capabilities and spectroscopic techniques, it is now possible to directly observe structural phase transitions and probe materials properties on the atomic scale [5,6]. Here, we describe our studies of the *1111*-type compound LaFeAsO using a battery of high-resolution and *in situ* TEM techniques in real (imaging), reciprocal (diffraction), and energy (spectroscopy) space. This material, which when electron-doped exhibits high- $T_c$  superconductivity, undergoes a crystallographic phase transition at 160 K from tetragonal to orthorhombic and also shows AF stripe ordering below 140 K [1,2]. To study the room-temperature tetragonal phase, we used an aberration-corrected FEI Titan G2 60-300 (S)TEM and an FEI Tecnai G2 F30 TEM to obtain atomic-resolution images, selected-area electron diffraction (SAED) patterns, and X-ray energy-dispersive spectroscopic (XEDS) profiles. For the orthorhombic and stripe-ordered AF phases, we used a Gatan 626 cryo-holder. In support of the experiments, we performed image simulations using a multislice method [7].

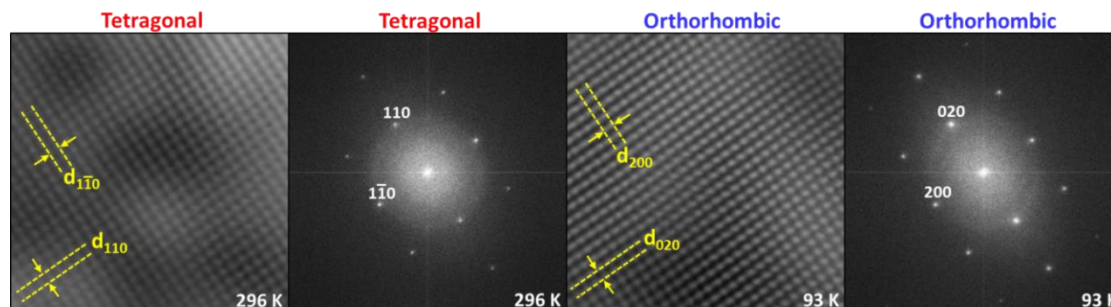
The combined imaging and diffraction experiments show that, regardless of the structural phase, dynamical effects on different regions of the same specimen suppress Z-contrast and produce qualitatively-similar scattering. For example, Figure 1 (left panel) shows a high-angle annular dark-field STEM (HAADF-STEM) image of (La,As) and (Fe,O) atomic columns as viewed along the [001] zone axis. It can be seen that the Z-contrast predicted to be present via image simulation is absent. Further, the right panel displays an SAED pattern along the [001] direction at 296 K showing a (100) reflection, whereas the same reflection is absent in the FFT of an atomic-resolution bright-field conventional TEM (CTEM) image also obtained at 296 K and, thus, the tetragonal phase. The implications for these and other results on directly observing the crystallographic phase transition in LaFeAsO via *in situ* TEM will be discussed [8].

## References:

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**Figure 1.** (Left Panel) Room-temperature HAADF-STEM image of LaFeAsO along the [001] zone axis. The LaFeAsO tetragonal crystal structure (top) and a simulated HAADF-STEM image (bottom) with Z-contrast are overlaid on the image with the atomic columns labeled. (Right Panel) SAED pattern showing the [001] orientation and presence of the (100) Bragg spot.



**Figure 2.** Atomic-resolution bright-field CTEM images and corresponding FFTs at 296 K (tetragonal) and 93 K (orthorhombic) from an LaFeAsO crystal viewed along the [001] zone axis. The  $d$ -spacings that give rise to spots in the observed FFTs are labeled. Note the absence of the (100) spot in the FFTs, regardless of temperature.