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In Situ Observation of Phase Separation in High-Temperature Superconductor $La_{2-x}Sr_xCuO_4$

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Since the remarkable discovery of superconductivity in 1911 [1], the research of achieving high-temperature superconductivity has been of interest for over a century. Cuprate oxide superconductor only occurred since 1980's because initial studies indicated low-temperature superconductivity, compared to intermetallic compounds [2]. However, these oxide superconductors have quickly become the most heavily studied material system because of its records of high-temperature superconductivity [2,3]. One challenge in high-temperature superconductivity is to realize the homogeneity and stability of a material [4,5].

In this work, in situ heating experiments of $La_{2-x}Sr_xCuO_4$ (x=0.03), as a representative high-temperature cuprate oxide superconductor, is demonstrated in a transmission electronic microscope. So far, there have been many reports about a well-known phase transition in $La_{2-x}Sr_xCuO_4$ system, which is the transition of the crystal structure from tetragonal to orthorhombic by CuO_6 octahedra tilts [6]. Here, on top of the phase transition, we demonstrate a unique phase separation phenomenon based on the analyses of in situ transmission electron microscopy (TEM) and electron energy-loss spectroscopy (EELS).

To obtain high-resolution scanning TEM (STEM) images, an aberration-corrected FEI Titan G2 60-300 STEM, operated at 300 keV, was used. Convergence semi-angle of the STEM incident beam was 24.3 mrad and high-angle annular dark-field (HAADF) images were recorded with detector angles of 41-200 mrad. Heated-stage TEM experiments was performed using a Gatan 652 double-tilt heating holder in an FEI Tecnai G2 F30 STEM with TWIN pole piece operating at 300 keV and equipped with a Gatan 4k×4k Ultrascan CCD. Bright-field TEM (BF-TEM) images and selected-area electron diffraction (SAED) patterns were acquired using the microscope at each temperature setting. EELS spectra were recorded using a Gatan Image Filter (GIF) spectrometer attached to the microscope.

A phase separation of $La_{2-x}Sr_xCuO_4$ was monitored by in situ heating experiments in TEM (vacuum level of ~10^{-7} Torr). The phase separation took place from as low as 150 °C; however, it occurred mostly in the temperature range of 350-450 °C (Figure 1). Two resultant phases were identified as metal Cu and distorted bixbyite La_2O_3 by SEAD pattern and EELS analyses. A similar phase decomposition was also reported in $Nd_{2-x}Ce_xCuO_{4\pm\delta}$ system [7]. The existence of the resulting phases after cooling indicates that the phase separation process is irreversible in our experiment setup. The EELS results demonstrated that bulk plasmon peak, O K and La $M_{4,5}$ edges change in the temperature range of 350-450 °C (Figure 2). The bulk plasmon peak from the distorted bixbyite La_2O_3 located at 25.2 eV and showed unique O K edge. We discuss the evolution of crystal structures and EELS peaks as function of temperature in detail [8].

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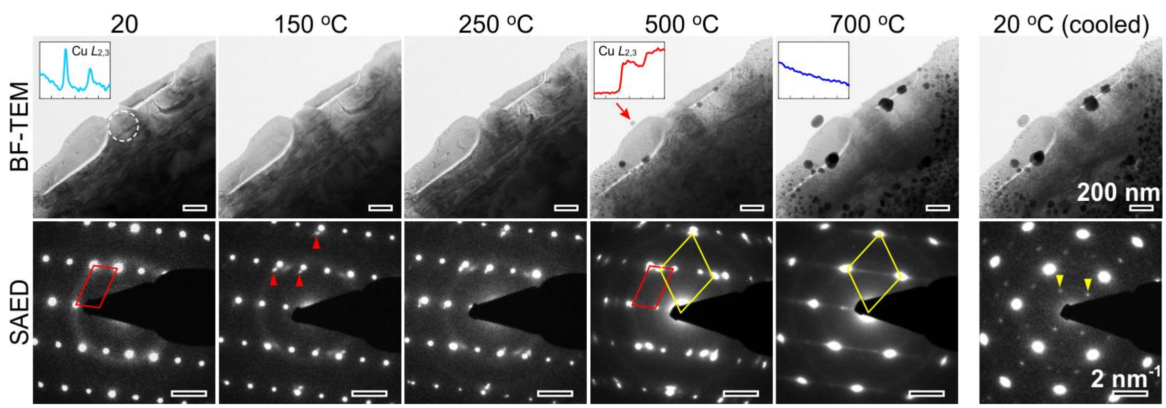

Figure 1. BF-TEM images (top panels) and corresponding SAED patterns (bottom panels) at selected temperatures during a heating experiment. Insets in panels at RT, 500 °C, and 700 °C are core-loss EELS spectra in the range from 910 to 970 eV of energy loss.

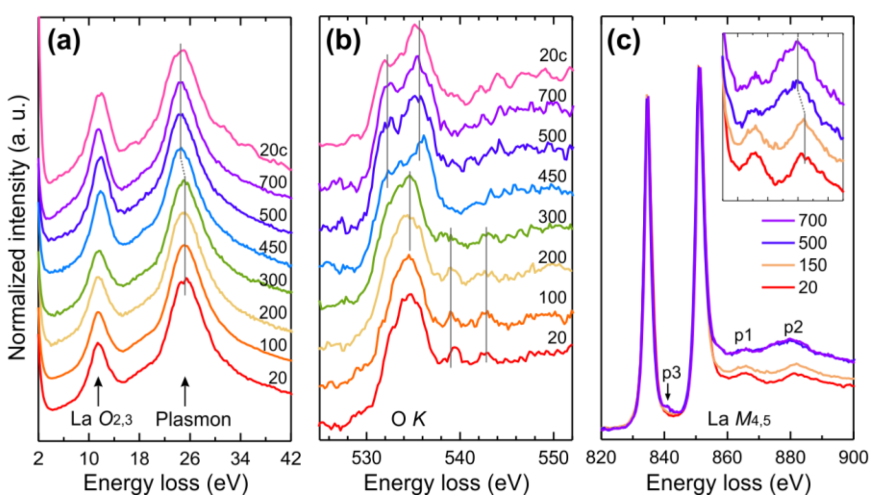

Figure 2. (**a**) Low-loss, (**b**) O *K* edge, and (**c**) La $M_{4,5}$ edge EELS spectra at selected temperatures (in °C) during a heating experiment. Insets in (**c**) shows magnified EELS spectra in the range of 855-895 eV of energy loss. The 20c represent the 20 °C after cooling.

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