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Imaging electronic phase separation in $La_{1-x}Ca_xMnO_3$ using electron nano-diffraction

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Doped manganites have attracted considerable interests because of their rich and complex physical phenomena occurring in these systems, for example, colossal magnetoresistance (CMR). Nanoscale phase separation has often been proposed but direct observation has so far been lacking. In this study we used electron nano-diffraction imaging (ENDI), an experimental technique combining structure sensitivity and nanometer scale spatial resolution, to map clusters as the specimen was taken through a phase transition. The principle of this technique involves scanning the electron probe on the sample and plotting the intensity of the superlattice reflections (SR) as a function of the probe position, as shown in Fig. 1.

The well known phase diagram of $La_{1-x}Ca_xMnO_3$ has shown that three phases dominate in a wide doping range at different temperatures: 1. The paramagnetic (PM) phase is favored at high temperature; 2. The ferromagnetic (FM) phase is favored at low temperatures for $0.2 < x < 0.5$; 3. The charge ordering (CO) phase is favored at low temperatures for $0.5 \le x \le 0.8$ [1]. Here we report the observation of the CO phase as nanometer-sized droplets during the PM-FM phase transitions at the doping range $0.33 < x < 0.50$, a regime where the CO phase has not been observed directly. Fig. 2a shows the electron diffraction patterns (DP) obtained from single crystal domains, at [010] zone axes with the *Pnma* notation $a \approx c \approx b / \sqrt{2}$, as the samples are taken across their transition temperatures. Diffuse SRs are clearly seen and indicate the presence of modulation structures similar to those of the CO phase [2]. The wave vectors of the SRs are found to be related to the doping levels and temperatures, as plotted in Fig. 2b.

The distribution of the CO clusters in $La_{0.55}Ca_{0.45}MnO_3$ obtained by ENDI is shown in Fig. 3 at different temperatures. The CO clusters, which are seen here for the first time, appear as nanometer-sized droplets and their density changes markedly during the phase transition. The direct observation of these nanoscale clusters brings a new opportunity to study nanoscale phase separation and interpret the mechanism of the phase transitions in $La_{1-x}Ca_xMnO_3$ [3].

References

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[3]. Research sponsored by the Division of Materials Sciences and Engineering, Office of Basic Energy Sciences, U.S. Department of Energy, under contract DE-AC05-00OR22725 with Oak Ridge National Laboratory, managed and operated by UT-Battelle, LLC.

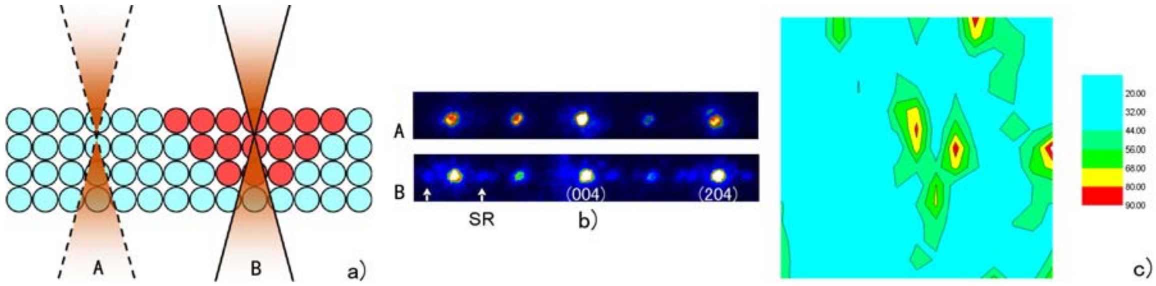

Fig. 1. A schematic diagram showing the principle of the electron nano-diffraction imaging (ENDI). Electron diffraction patterns (DP) were obtained by scanning a nano-meter-sized electron beam on the sample. When the electron beam sits on an area with structural modulations, the DP has the diffuse superlattice reflections (SR) (see B). c) is an example of the contour map constructed by plotting the intensity of the SRs obtained from DPs. The flat area has the intensity below the noise level.

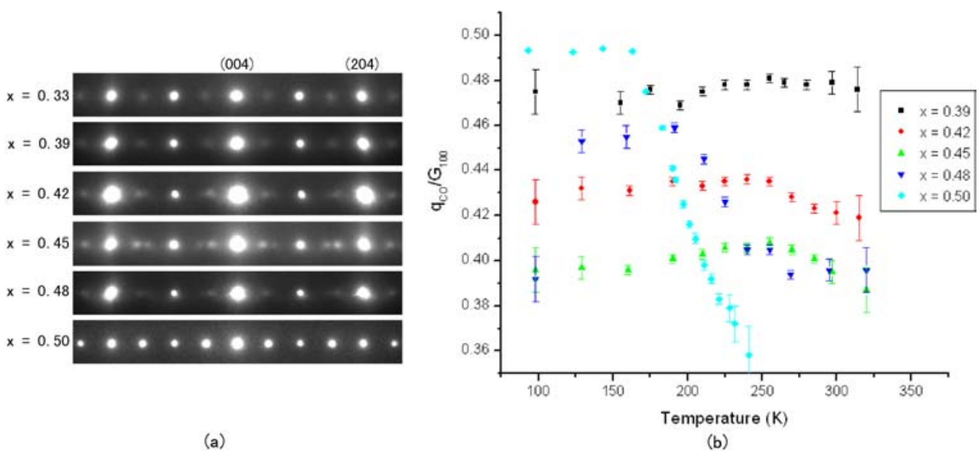

Fig. 2. (a). Systematic row electron diffraction patterns (DP) from $La_{1-x}Ca_xMnO_3$ samples at x = 0.33, 0.39, 0.42, 0.45, 0.48, and 0.50 at T = 200 K. The DPs contain the (004) and (204) reflections as well as diffuse superlattice reflections. (b). The measured wave vector q_{CO} from the DPs were plotted versus temperature for all the samples.

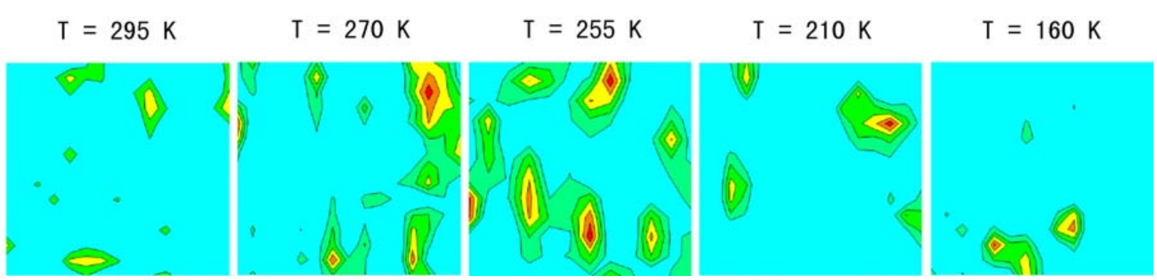

Fig. 3. Evolution of the charge ordering phase in $La_{1-x}Ca_xMnO_3$ samples at x = 0.45 showing the nanoscale phase separation at different temperatures. Each image is mapped from the electron nano diffraction pattern and is 12 × 12 nm^2 in size.

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