In-situ TEM Electrical Biasing of LAO/STO Interface-Devices Revealing Charge Modulation and Associated Structural and Chemical Changes

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The recent discovery of a two-dimensional electron gas (2DEG) at the interface between insulating perovskite oxides $SrTiO_3$ (STO) and $LaAlO_3$ (LAO) was made possible by advances in atomic layer-controlled growth. It has been demonstrated that the 2DEG is localized within a few nm of the interface, and that the carrier concentration can be altered with an electric field and/or lattice strain. Controlling such properties of 2DEG by applying external biases has been demonstrated in various prototype device structures such as field effect transistor, diode and nonvolatile memory devices. Eventually, seeing such changes in-situ during the operation of devices can improve our understanding and also provide a technical breakthrough for further optimization of devices.

In this study, in-situ inline electron holography biasing experiments have been carried out on the epitaxially grown LAO (10 u.c.)/STO (15 u.c.)/LAO (3 u.c.) devices to observe charge density modulation by electrostatic gating. TEM samples for in-situ biasing experiment were prepared by using focused ion beam (FIB) (Fig. 1). A thin lamella was lift out and fixed on a Si MEMS chip and then further milled for electron transparency in a FIB using a low energy Ga^+ ion beam. For the application of electric field across the LAO/STO interface, electrical circuit was constructed by cutting trenches for electrical isolation of one electrode from each contact. While negative/positive DC biases are applied to the SRO top electrode, inline electron holography, STEM imaging and EELS have been carried out in-situ in TEM (GRAND ARM 300 CF).

The in-situ TEM *I-V* curves of the device show no significant hysteresis in voltage ramping but a clear change in the resistance at +0.5 V and -0.3 V, very similar to a typical varister. We simulated the *I-V* characteristics for this device structure by using density functional theory and found that a resonance tunneling type conduction takes place, in good agreement with the in-situ TEM result. The potential profile obtained by inline holography at each voltage shows a similar tendency as a resonance tunneling device would exhibit. The charge density maps clearly show a deep valley near the upper interface LAO/STO, which corresponds to the existence of 2DEG. The charge density of 2DEG measured at each voltage in shows an increase and decrease of the 2DEG density at positive and negative voltage, respectively (Fig.2). We also observed a noticeable change the lattice strain in in-situ STEM HAADF images. The out-of-plane strain maps obtained by geometric phase analysis (GPA) of STEM HAADF images show the lattice expansion of STO layer near the upper interface along the out-of-plane direction at both positive and negative voltages. In addition to the strain analysis, the valence state analysis by in-situ EELS will be discussed in greater details.

Reference

[1] H.Lee, et.al., *Nature Materials* (2018)
[2] K.Song, et.al., *Nature Nanotechnology* **2** (2018), 1400281

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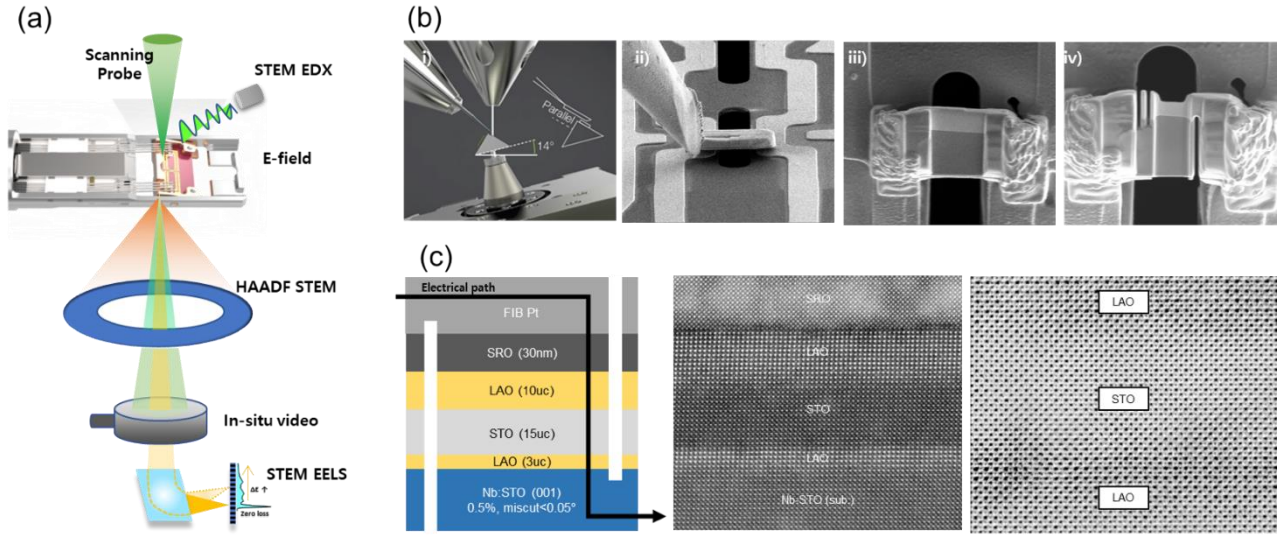

Figure 1. TEM sample preparation for in-situ biasing of switchable 2DEG systems. (a) TEM biasing holder. (b) TEM image of a TEM sample prepared by FIB. (c) Schematic showing the electrical contact of the device. The top SRO electrode together with Pt layer is connected to the left metal pad and the Nb-STO substrate to the right metal pad by cutting trenches for electrical isolation of one electrode from each contact. (d) FIB procedure for TEM sample preparation. (e) STEM HAADF and ABF images of the LAO/STO interface.

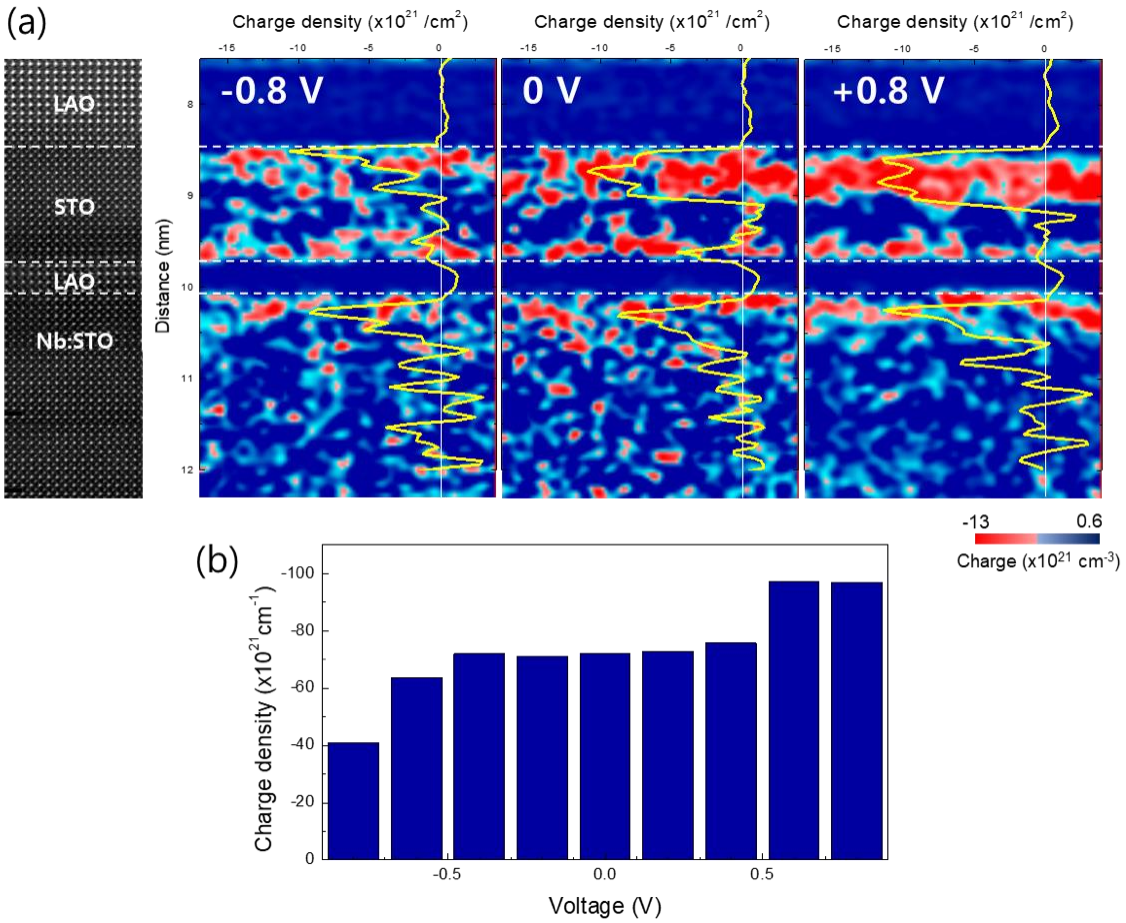

Figure 2. Charge density maps obtained by in-situ inline electron holography. (a) Charge density map derived from the phase shift map obtained by inline holography at -0.8 V, 0 V and +0.8 V. (b) Integrated charge density near the upper LAO/STO interface where the 2DEG exists.