Supporting Information

Atomically Resolved Electronic States and Correlated Magnetic Order at Termination Engineered Complex Oxide Heterointerfaces

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Supporting information 1

1. The height histogram of Nb:$SrTiO_3$/ $La_{0.7}Sr_{0.3}MnO_3$/ $BiFeO_3$

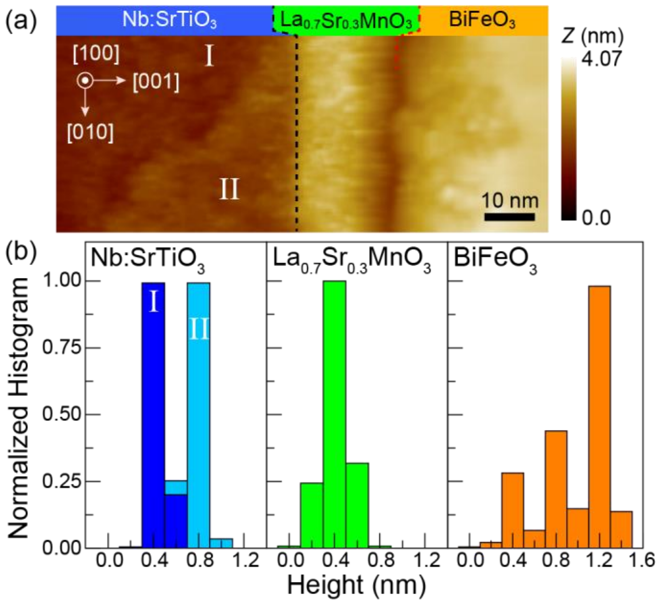

Figure S1: *(a) STM image of the cross-sectionally cleaved Nb:$SrTiO_3$/ $La_{0.7}Sr_{0.3}MnO_3$/ $BiFeO_3$ surface and (b) corresponding height histograms of the Nb:$SrTiO_3$, $La_{0.7}Sr_{0.3}MnO_3$, and $BiFeO_3$ surface areas, respectively.*

The STM images and the corresponding height histograms of the Nb:$SrTiO_3$ substrate and the $La_{0.7}Sr_{0.3}MnO_3$ film (**Fig. S1**) show that both cleavage surfaces exhibit atomically flat surfaces with only one or two monolayers exposed. In contrast, the roughness of the $BiFeO_3$ cleavage surface is much higher with typically three to four monolayers exposed (see height histogram in **Fig. S1(b)**). The increased roughness of the $BiFeO_3$ (100) surface is attributed to the presence of a spontaneous polarization along the [111] direction in $BiFeO_3$, which induces at the initially non-polar (100) plane a partially polar character. The polar character increases the cleavage energy for the separation of charges. This typically reduces the cleavability and hence induces a larger roughness of the resulting cleavage surfaces.

Supporting information 2

2. Averaged spectra of the normalized differential conductivity in Nb:$SrTiO_3$/ $La_{0.7}Sr_{0.3}MnO_3$/ $BiFeO_3$ heterostructures

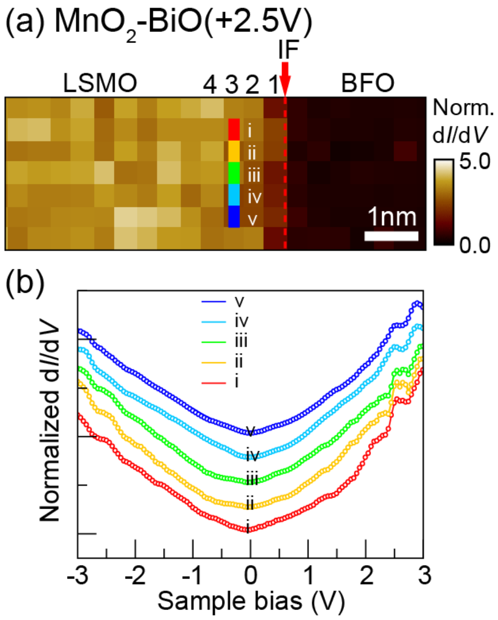

***Figure S2**: (a) Normalized dI/dV image close to the MnO_2-BiO terminated interface. The position of the MnO_2-BiO interface is indicated by "IF", and the numbers 1, 2, 3, 4 are representative the positions away the interface by 1, 2, 3, or 4 layers. The separation of each number is 0.4 nm. (b) The positions labeled i, ii, iii, iv, and v are at the same distance from the "IF" in (a). (b) Raw normalized dI/dV curves as acquired at the positions i to v. Their average has been shown in the manuscript for the spatial position labeled 3 perpendicular to the interface.*

Each normalized d*I*/d*V* curve shown in Figure 3 in the manuscript is the average of 5 raw data curves. For example, the normalized d*I*/d*V* curve labeled number 3 of the $La_{0.7}Sr_{0.3}MnO_3$ thin film is the average of the raw d*I*/d*V* curves labeled i, ii, iii, iv, and v shown in **Figure S2**. The overall quality of the raw data and the averaging leads to rather smooth d*I*/d*V* curves shown in Figure 3.

Supporting information 3

3. Physical origin and subtraction procedure of the background in STS spectra

The background in STS spectra originates from the dependence of the tunneling barrier height on the applied bias voltage (i.e. energy). This can be best explained when recalling the transmission coefficients [*D(W)*] in the WKB approximation, given by $D(W)= exp\{-2(m/\hbar^2)^{1/2}\int[V(z)-W]^2\,dz\}$, with W being the normal component of the energy [$W=E - \hbar^2 k_{ll}{}^2/(2m)$], E the total energy, k_{ll} the component of wave vector parallel to the interface, and m the effective mass. $V(z)$ is the (trapezoidal) potential barrier within the vacuum between the sample surface and the tip [1]. In this approximation the current density in STM, given by $J=(e/4\pi^2\hbar^2)\int D(E)\,dE$, yields a tunneling current (I), a differential conductance (dI/dV), and a normalized differential conductivity as in **Fig. S3**.

Comparing the experimental normalized conductivity spectra in **Fig. S4** with that calculated on basis of the transmission probability only (**Fig. S3c**), it becomes obvious that the background follows very closely the voltage dependence originating from the transmission probability, which is indeed the physical origin of the background in the STS spectra.

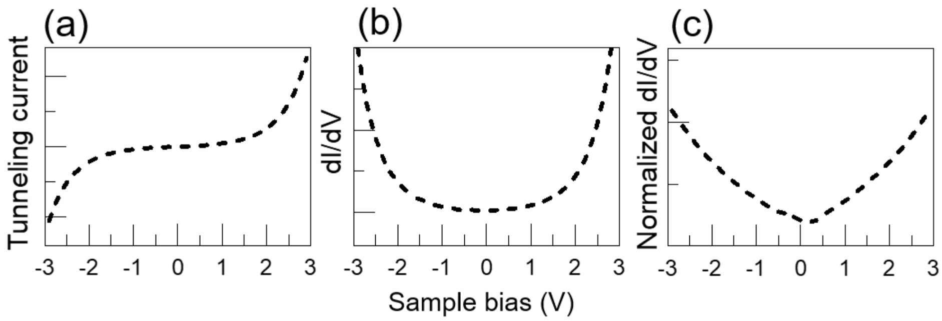

Figure S3: *Simulation of (a) the tunneling current (I), (b) the differential conductance (dI/dV), and (c) the normalized differential conductivity using the transmission probability only.*

Next, we explain the background subtraction procedure using one dI/dV curve of those shown in **Fig. S4**. The normalized conductivity of this example spectrum is shown in **Fig. S4**(a) as green line. To emphasize the characteristic peak, the dI/dV signal is magnified in **Figure S4**(b). In order to obtain the shape of the background we recall that in the present STS measurement, the work function of metallic tip (Φ_m) is 4.5 eV, while the work function of sample (LSMO) (Φ_s) is 4.8 eV [2]. The tip-sample separation (s) is 1.0 nm. Furthermore, the hole and electron effective mass of LSMO are 6.0 and 4.0, respectively [3]. With these values a normalized differential conductivity based solely on the transmission probability can be derived as that shown in **Fig. S3**(c). Its shape can be fitted to the background resulting in the black dashed curve in **Fig. S4**(b). Subtracting the such determined background results in the characteristic peak only shown in **Fig. S4**(c). Through this procedure, a reliably energy position of the peak can be determined and thereby the evolution of the peak energy as a function of the spatial distance from the interface.

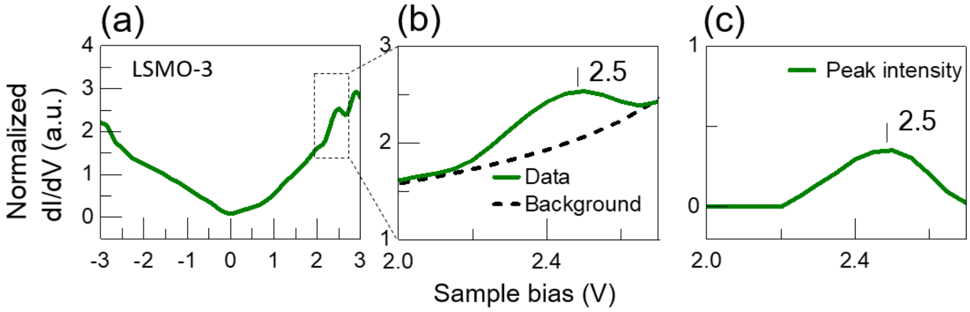

Figure S4: *The background subtraction procedure: (a) Overview experimental normalized differential conductivity spectrum, (b) magnified peak in the normalized differential conductivity spectrum with fitted background with a shape arising from the energy dependence of the transmission coefficient, and (c) peak after background subtraction from the original dI/dV signal.*

Ref：

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[2] Fenghong Li *et al*., J. Phys. Chem. C **115**, 16947 (2011).

[3] Leng Liao *et al*., Phys. Stat. Sol. A **206**, 1655 (2009).