Supporting Information

Step Terrace Tuned Anisotropic Transport Properties of Highly Epitaxial LaBaCo$_2$O$_{5.5+\delta}$ Thin Films on Vicinal SrTiO$_3$ Substrates

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1. X-ray Diffraction from all vicinal substrates

Figure S1 shows the X-ray diffraction data from the as-grown LBCO thin films on 0.5° , 3°, 5° miscut (001) STO substrates, respectively. No significant difference was observed in the patterns from all vicinal surfaces. The as-grown films are of a pure LBCO phase with the *c*-axis highly normal to the substrate surface and with only (00*l*) peaks present from the LBCO film overlapped with STO substrate.
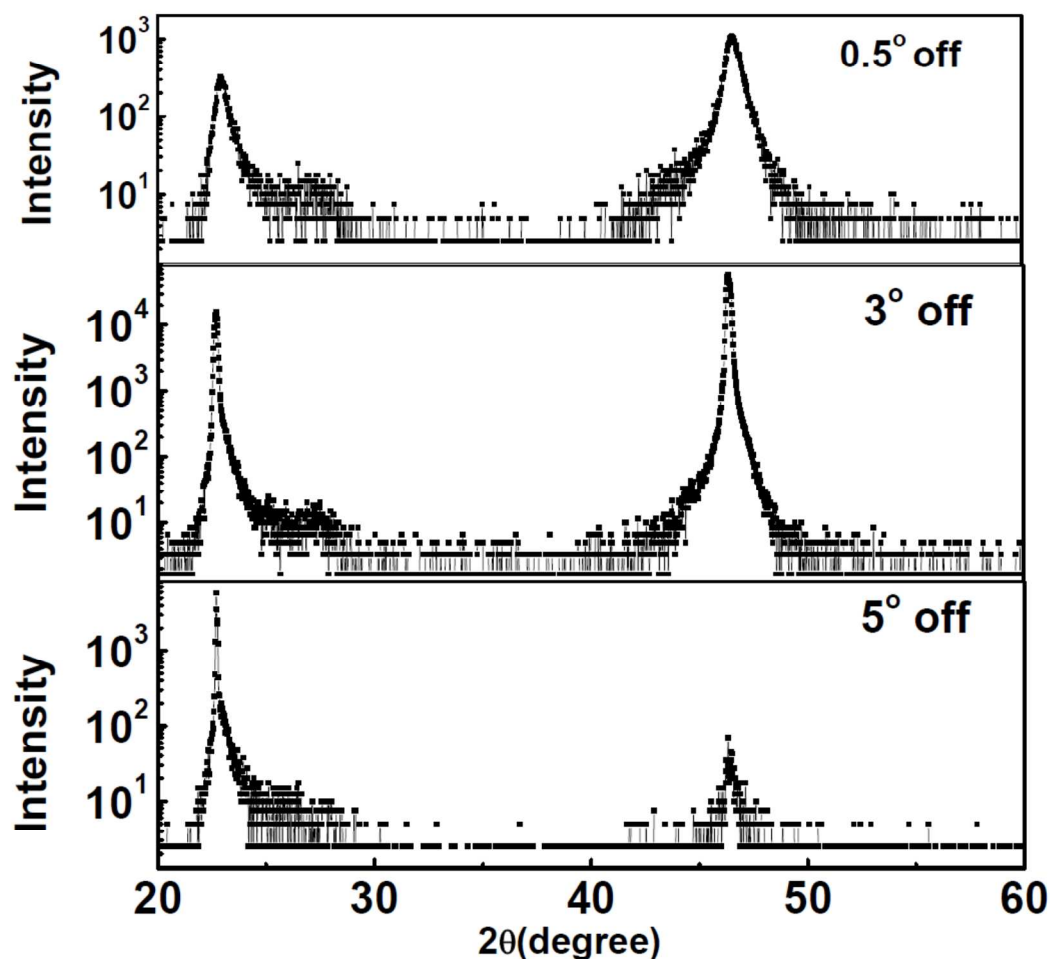

Figure S1. The X-ray diffraction data from the as-grown LBCO thin films on 0.5° , 3°, 5° miscut (001) STO substrates, respectively.

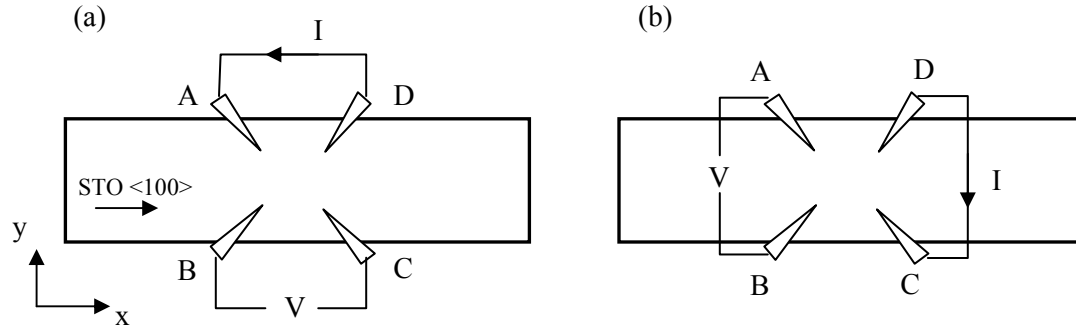## 2. Four-probe electrical transport measurement

Figure S2. Schematic illustration of setups for four terminal method used in thin film electrical resistance measurement. (a), The Current is applied between probe A and D, and voltage is measured between probe B and C. The nominal resistance (slope of V-I curve) is obtained as R_x. (b), Current is applied between probe C and D, and voltage is measured between probe A and B. As-obtained resistance is denoted as R_y. STO<100> direction, parallel to the miscut step gradient direction, is shown in (a).

The square four-terminal method was used for measuring the resistance of thin films (1). In this method four probes are placed in a square shape, equally spaced, as shown in Figure S2. Sweeping current is applied on one pair of adjacent electrodes, and the voltage potential is measured on the other pair of electrodes by using of Keithley 4200. Thus, the nominal resistance (R_x and R_y) can be obtained from the slopes of the V-I curves along orthogonal measurement directions.

3. Transport Property Analysis

We analyzed the electronic transport properties with both the thermal activation model and the small polaron model.

1) The thermal activation model (ln R ∝ 1/T)

The Arrhenius Equation of the thermal activation model is as follows: $R = Ae^{(-Ea/kT)}$. Here, R is the resistance of the LBCO thin film, A is a constant, Ea is the activation energy in electron volts (eV), k is Boltzmann's constant (8.6×10^{-5} eV/K), and T is the absolute temperature. With the thermal activation model, the fitted activation energies of the LBCO films were determined to be 31.7 and 29.1 meV, 27.5 and 27.1 meV, and 13.1 and 8.9 meV on the 0.5°, 3.0° and 5.0° miscut substrate, respectively.
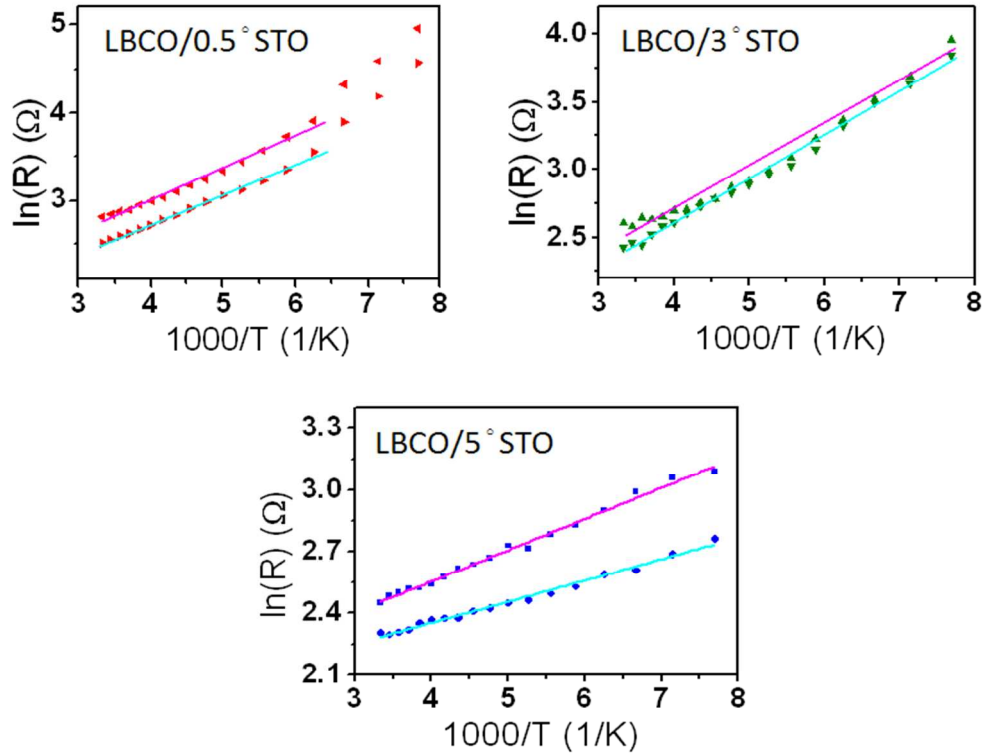

Figure S3. The analysis of the electronic transport properties with the thermal activation model (ln R ∝ 1/T).

2) The small polaron model (ln R/T ∝ 1/T)

The formula of small polaron model can be expressed as:

$$R = C_1 k_B T exp(\frac{\varepsilon_3}{k_B T})$$

The small polaron mechanism shows much better fitting than the thermal activation model, suggesting that in the LBCO thin films, the interaction between electron and lattice is strongly localized. The activation energies of the LBCO films were determined to be 57.5 and 54.4 meV, 44.3 and 43.1 meV, and 29.7 and 25.6 meV on the 0.5°, 3.0° and 5.0° miscut substrate, respectively.
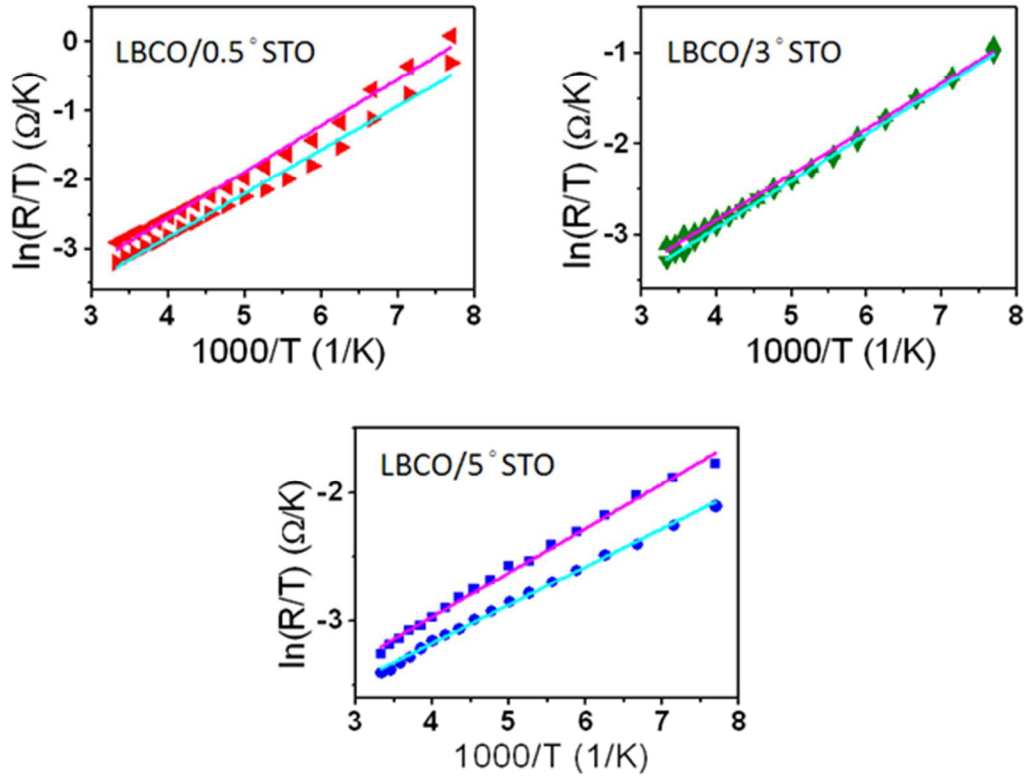

Figure S4. The analysis of the electronic transport properties with the small polaron model (ln R/T ∝ 1/T).