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Tunability of conduction at the $LaAlO_3/SrTiO_3$ heterointerface: Thickness and compositional studies

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The role of chemistry, film thickness, and oxygen pressure in influencing the electrical and thermal transport properties of $LaAlO_3/SrTiO_3$ heterointerfaces is explored. Unit-cell precise growth was accomplished for films between 3 and 160 unit cells thick using reflection high-energy electron diffraction-assisted pulsed-laser deposition. Subsequent temperature-dependent studies of electrical resistivity reveal three important observations: (1) by tuning the laser fluence, we can systematically tune the interfacial conductance in a step-wise manner in this system, (2) all films exhibit a critical thickness of 3–4 unit cells for the onset of conduction, and (3) the nature of the conductance is highly influenced by the stoichiometry of the $LaAlO_3$ film with La-deficient samples showing dramatic changes with thickness, while stoichiometric and La-excess films show little dependence. Time-domain thermoreflectance studies show a diminished interfacial thermal conductance for the La-deficient films when compared to La-excess and stoichiometric films, suggesting that the interfacial conductance is more influenced by extrinsic factors such as oxygen deficiency. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4896778]

Considerable work has focused on the conducting interface between $LaAlO_3$ and $SrTiO_3$[1] and the origin of this so-called 2-dimensional electron gas has been hotly debated.[2] A number of interesting findings related to these heterointerfaces have been made including the observation of magnetism,[3] superconductivity,[4] built-in polarizations,[5] and more.[6,7] Several mechanisms have been proposed to explain these observations, including electronic reconstruction,[8–10] oxygen vacancy defects,[11–13] knock-on damage from high-energy adatoms,[14] and interfacial intermixing of cations.[15–17] Although a number of reports support the idea that electronic reconstruction certainly plays a role as an intrinsic mechanism for conduction,[18] in practice, the aforementioned extrinsic effects may also factor in and it can be difficult to differentiate such effects. This is especially problematic for high-energy growth processes such as pulsed-laser deposition;[19] one of the primary deposition techniques for complex oxides.

More recently, several studies have begun to investigate cation stoichiometry as an additional source of extrinsic effects. It has been demonstrated that there is a strong link between $LaAlO_3$ cation stoichiometry and the $LaAlO_3/SrTiO_3$ interfacial properties. For instance, the critical thickness for the onset of conductance can be tuned by growing $(LaAlO_3)_{1-x}(SrTiO_3)_x$ films with various values of x (Ref. 20) and one can systematically tune the interfacial transport by altering the La/Al ratio of the $LaAlO_3$ film.[21–23] This Letter builds upon such studies to report on the combined effects of film stoichiometry, thickness, and oxygen pressure during growth in influencing the electrical and thermal transport properties of $LaAlO_3/SrTiO_3$ heterointerfaces. This work demonstrates that the electrical resistivity can be tuned, in a step-wise manner, using laser fluence, that all films (grown at sufficiently oxidizing conditions) exhibit a critical thickness of 3–4 unit cells for the onset of conduction and that the nature of the conductance is highly influenced by the stoichiometry of the $LaAlO_3$ film with La-deficient samples showing dramatic changes with thickness, while stoichiometric and La-excess films show little dependence. Time-domain thermoreflectance studies show a diminished interfacial thermal conductance for the La-deficient films when compared to La-excess and stoichiometric films, suggesting that the interfacial conductance is more influenced by extrinsic factors such as reduction of the underlying substrate.

Films of $LaAlO_3$ with thicknesses between 1.13 nm (3 unit cells, uc) and 60 nm ($\sim$160 uc) were grown via pulsed-laser deposition following established procedures.[21] Briefly, a laser frequency of 1 Hz was used for all growths resulting in a growth rate of $\sim$12 pulses/uc. The laser fluence was varied between 1.2 and 2.5 J/cm^2 by changing the laser spot size from 0.043 to 0.071 cm^2, while holding the laser energy constant (85 mJ). All growths were completed at a temperature of 750 °C as measured via optical pyrometry in oxygen pressures of 10^{-6} Torr or 10^{-3} Torr. All films were grown on single crystal $SrTiO_3(001)$ substrates treated via standard methods.[24,25] Following growth, films were cooled at

5 °C/min to room temperature at the growth pressure. For films between 1.13 and 3.79 nm (3–10 uc) in thickness, *in situ* monitoring of the growth process with reflection high-energy electron diffraction (RHEED) was completed to assure unit-cell level control of the heterostructures. A sample RHEED pattern is provided along with the tracked intensity oscillations for a characteristic growth of a 3.79 nm (10 uc) thick film [Fig. 1].

Following our established procedure,[21,26,27] an array of techniques were used to probe the chemical, structural, electrical, and thermal properties. Consistent with prior work,[21] X-ray diffraction studies (regardless of the laser fluence and film thickness) reveal single-phase and epitaxial $LaAlO_3$ films. Subsequent atomic force microscopy studies reveal smooth, atomic-level terraced, island-free films. Study of film chemistry, however, reveals deviations in the cation stoichiometry as the laser fluence is changed. Using X-ray photoelectron spectroscopy and Rutherford backscattering spectrometry, we observe that $LaAlO_3$ films grown at 1.2 J/cm^2 exhibit a 4%–5% excess of La (as calculated by [La]/([La] + [Sr]) × 100), films grown at 1.6 J/cm^2 exhibit a nearly ideal stoichiometry, and films grown at 2.0 J/cm^2 exhibit a 4%–5% deficiency of La.

To demonstrate the potential of chemical control in $LaAlO_3/SrTiO_3$, we have gone on to exert fine-level control over the growth of the $LaAlO_3$ thin films. Focusing on La-deficient films as an example, by varying the laser fluence between 1.7 and 2.5 J/cm^2, we can produce step-wise changes in the interfacial conductance. Temperature-dependent sheet resistance studies, performed in a van der Pauw configuration, were completed on 3.79 nm (10 uc) thick films of $LaAlO_3$ grown at 10^{-3} Torr O_2 that were controlled to possess 1%, 2%, 3%, 4%, and 10% La-deficiency [Fig. 2] (La-deficiency values are ascribed based on chemical studies and should be considered accurate within +/−0.8%). In all cases, the heterointerfaces exhibit metallic-like conductivity from room temperature downward. In some of the heterostructures (i.e., 1%, 2%, 3%, and 4% La-deficient), the transport undergoes a crossover from metallic- to insulator-like transport at a critical temperature which decreases with increasing La-deficiency. This change in the crossover transition temperatures suggests a potential change in the nature of the interfacial conductance (trending towards 3-dimensional conductance with increasing La-deficiency). This is supported by the fact that as the La-deficiency is increased, the transport trends towards that of a reduced $SrTiO_3$ substrate (annealed at 10^{-6} Torr and 750 °C for the same time it takes to create a 3.79 nm (10 uc) thick film), which exhibits 3-dimensional transport. We believe that this could potentially be correlated to a cation (non)stoichiometry driven reduction of the substrate which is exacerbated by increased La-deficiency (or Al-excess). This will be expanded upon more later.

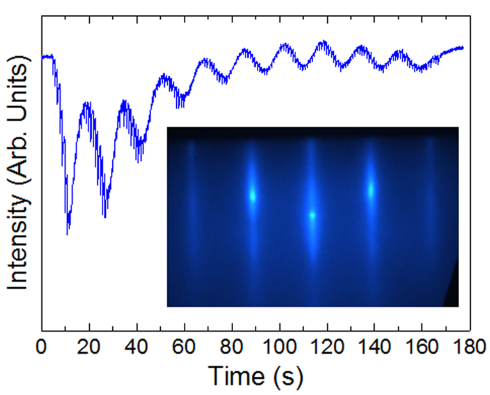

FIG. 1. RHEED-assisted growth of a 10 unit cell thick $LaAlO_3$ films on a $SrTiO_3$(001) substrate. Clear oscillations in the intensity of the specular RHEED spot reveal layer-by-layer growth (where each individual laser pulse is evident within the oscillations). The inset shows the RHEED pattern immediately following growth and is indicative of a very smooth film surface.

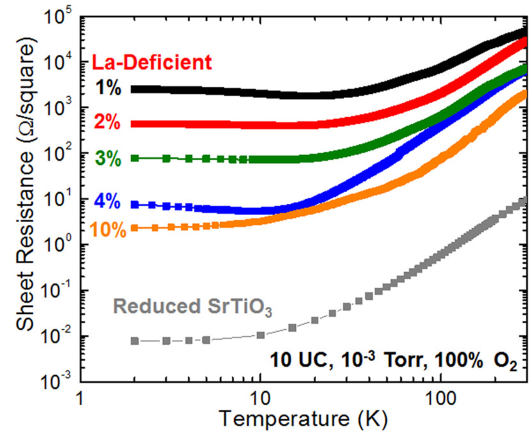

FIG. 2. Sheet resistance as a function of temperature for a series of 10 unit cell $LaAlO_3/SrTiO_3$(001) heterostructures, where the $LaAlO_3$ was controlled to possess 1%, 2%, 3%, 4%, and 10% La-deficiency. Increasing La-deficiency in the film leads to reduce crossover temperature from metallic-like to insulator-like conductivity and to an overall reduction in sheet resistance and a trend towards three-dimensional conduction like that observed in a reduced $SrTiO_3$(001) substrate.

We have gone on to study the thickness dependence of the interfacial conductance by probing the transport in 1.13 (3), 1.52 (4), 1.90 (5), and 3.79 nm (10 uc) thick films, grown at 10^{-3} Torr and controlled to possess 4%–5% La-deficient [Fig. 3(a)], nearly stoichiometric [Fig. 3(b)], and 4%–5% La-excess [Fig. 3(c)] stoichiometries. Beginning with the La-deficient films, all films with thicknesses >1.13 nm (3 uc) exhibit a metallic, highly conducting transport profile, while growth of a 1.13 nm (3 uc) thick film produced an insulating interface. Interestingly, the sheet resistance of the conducting interfaces was found to scale inversely with thickness (i.e., thicker films yield lower sheet resistance). Similar studies of nearly stoichiometric films, again reveal that all films with thicknesses >1.13 nm (3 uc) possess metallic-like conductivity from room temperature down to 20–50 K, where they experience a crossover from metallic- to insulator-like conduction. Note that the transport profiles do not change appreciably with increasing film thickness and that the overall sheet resistance values are closer to that of the thinnest conducting La-deficient films. Once again, 1.13 nm (3 uc) thick, nearly stoichiometric films are found to be insulating. Finally, similar studies of the La-excess films again reveal that all films with thicknesses >1.13 nm (3 uc) exhibit interfacial conductance with metallic-like conductivity persisting down to ∼55 K, where they also undergo a crossover from a metallic- to insulator-like conductivity and, similar to nearly stoichiometric films, the transport profiles for films of

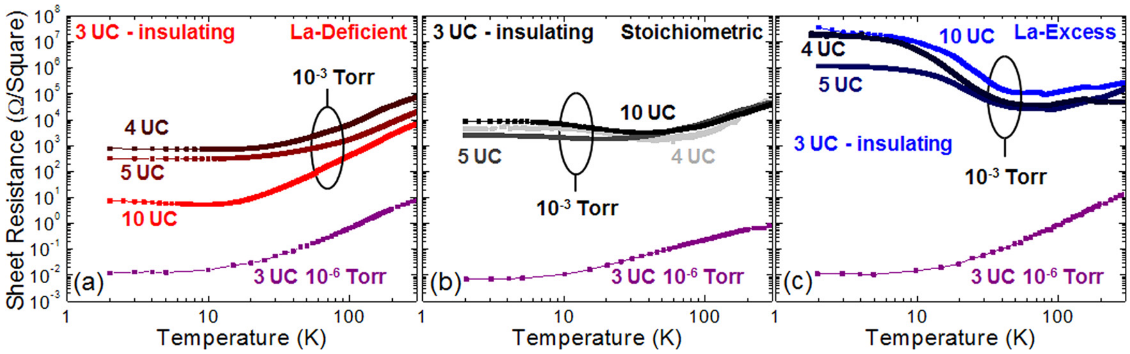

FIG. 3. Sheet resistance as a function of temperature and film thickness (3, 4, 5, and 10 uc) for $LaAlO_3/SrTiO_3(001)$ heterostructures, where the $LaAlO_3$ was controlled to possess (a) 4%–5% La-deficiency, (b) nearly stoichiometric, and (c) 4%–5% La-excess film stoichiometry at a growth pressure of 10^{-3} Torr of oxygen. For comparison, the sheet resistance of a film of like stoichiometry, but grown at 10^{-6} Torr (purple data), is provided in each panel.

various thicknesses are similar and no systematic trends are observed with changing thickness. Finally, 1.13 nm (3 uc) thick La-excess films are also observed to possess insulating behavior. Changing the growth pressure, however, to 10^{-5}–10^{-6} Torr (here, we show the data for only 10^{-6} Torr), one can observe highly conducting heterostructures even when the film thickness is only 1.13 nm (3 uc) thick regardless of the film stoichiometry [purple data, Fig. 3].

There are several important points to be made. First, all films grown at 10^{-3} Torr had an onset of measureable conductivity between 3 and 4 uc of thickness, regardless of stoichiometry. This may indicate that for all compositions, a polar-catastrophe-driven electronic reconstruction plays a role in the observed conduction. Second, for both the nearly stoichiometric and La-excess films, there is no strong correlation between film thickness and sheet resistance and this, together with the critical thickness for conduction, is suggestive that the observed interfacial conductance arises primarily from the *intrinsic* polar-catastrophe-induced mechanism. This stands in stark contrast to the effects observed in the La-deficient films, where the sheet resistance is observed to decrease with increasing film thickness (trending towards that of a reduced $SrTiO_3$ substrate). This is consistent with a number of prior works on non-stoichiometric $LaAlO_3$[21–23] and oxygen-deficient alumina ($Al_2O_{3-\gamma}$) films,[28,29] suggesting that defective films can potentially induce extrinsic conductivity in the $SrTiO_3$ substrate (separate from the polar-catastrophe-driven mechanism) as a result of reduction. Given that recent calculations[30] have shown that $LaAlO_3/SrTiO_3$ heterointerfaces with La-deficiency can have higher densities of La- and O-vacancy Schottky pairs and that Al possesses a large free energy of oxidation, it is reasonable to expect La-deficient (or Al-excess) films to induce oxygen reduction in the $SrTiO_3$. Thus, as the $LaAlO_3$ film is grown thicker and the substrate is subjected to reducing conditions for longer periods of time, it is reasonable to expect the substrate to become increasingly reduced and the interfacial sheet resistance values should decrease. This said, the La-deficient films do exhibit an onset for interfacial conductivity at thicknesses between 3 and 4 uc, suggesting that the polar catastrophe is likely present even in these interfaces, but that additional conduction in the substrate is activated by the reduction of the $SrTiO_3$, which ultimately overwhelms the interfacial conductance (especially at low temperatures, where the intrinsic response is found to exhibit a turn-up in resistance). This is also the case in the films grown at 10^{-6} Torr, where highly conducting behavior is observed even in 3 uc thick films.

These observations, in turn, suggest that $LaAlO_3/SrTiO_3$ interfaces with La-deficient films should exhibit an increased defective nature as compared to the nearly stoichiometric and La-excess films. To assess this concept, we have performed time-domain thermoreflectance (TDTR)[31,32] studies of the thermal conductance of the heterostructures for 4%–5% La-deficient, nearly stoichiometric, and 4%–5% La-excess films with thicknesses between 3.79 and 60 nm. Such TDTR studies are very sensitive to the cation stoichiometry and defect structures in epitaxial complex oxide films and thus provide another probe of the disorder at and near the heterointerface. The TDTR studies were completed by depositing a 80 nm thick Al transducer layer on the $LaAlO_3$ films and were carried out using a 10.3 μm beam diameter, 9.8 MHz modulation frequency, 16 mW pump power, and 8 mW probe power (consistent with prior studies).[26,27] After collecting the raw TDTR data for the $Al/LaAlO_3/SrTiO_3$ heterostructures, we compared this data with a thermal model using a one-dimensional diffusion equation.[33] We treat unknown thermal properties (in this case the $LaAlO_3$ thermal conductivity and the $LaAlO_3/SrTiO_3$ interfacial thermal conductance) as free parameters, which are adjusted to minimize the differences between our model and the raw data. There are two different ways that we can solve for our unknown thermal properties, depending on the thickness of the film. For films >15 nm, we model the following thermal resistances: (1) $Al/LaAlO_3$ interfacial thermal conductance, (2) $LaAlO_3$ thermal conductivity, (3) $LaAlO_3/SrTiO_3$ interfacial thermal conductance, and (4) $SrTiO_3$ thermal conductivity. After we complete this analysis, we obtain values for the $LaAlO_3$ thermal conductivity and the $LaAlO_3/SrTiO_3$ interfacial conductance for our three different compositions (consistent with prior work[27] on thick films). On the other hand, for thin films (i.e., 3.79 nm, 7.58 nm, and 15.2 nm thick films), we cannot uniquely solve for both the $LaAlO_3$ thermal conductivity and the $LaAlO_3/SrTiO_3$ interfacial thermal conductance because the measurement is not very sensitive to the thermal conductivity of the $LaAlO_3$ layer, and it contributes very little to overall thermal resistance of the system (see supplementary material for details on thermal sensitivity).[34] Thus, we combine these values into one "effective interfacial thermal conductance" which is largely dominated by the interfacial thermal resistance. In this case, our model considers the following thermal resistances: (1) Al/$LaAlO_3$ interfacial thermal conductance, (2) $LaAlO_3/SrTiO_3$ interfacial thermal conductance, and (3) $SrTiO_3$ thermal conductivity.

Since we have extracted two different data sets for thick and thin films, it is necessary to perform some additional data processing in order to directly compare the results. Our goal is to be able to probe the thermal properties of the $LaAlO_3/SrTiO_3$ interface, but as we discussed already, we cannot uniquely determine this in the thin films. In order to directly compare between both the thick and thin films, we plot the *total effective conductance* of each sample as a function of film thickness. The total effective conductance, which we call G, is given by $G^{-1} = G_I^{-1} + G_F^{-1}$, where G_F is the thermal conductance of the $LaAlO_3$ film and G_I is the thermal conductance of the $LaAlO_3/SrTiO_3$ interface. Since we have modeled our thin films as only interfaces, we can consider ${G_F}^{-1}$ to be zero and thus $G = G_I$.

When we plot G as a function of film thickness for the La-deficient, nearly stoichiometric, and La-excess films clear differences emerge [Fig. 4]. To better understand the effects, we model the effective thermal conductance for the film/interface system, as a function of film thickness, using the expression $G = \frac{g\left(\frac{\lambda}{h}\right)}{g+\frac{\lambda}{h}}$, where g is the interfacial thermal conductance between $LaAlO_3$ and $SrTiO_3$, λ is the thermal conductivity of the $LaAlO_3$ film, and h is the thickness of the $LaAlO_3$ film.[35] Using this model, we explore three hypothetical systems with different values for the thermal conductivity and the interfacial thermal conductance: (1) high interfacial thermal conductance (assumed to be 350 MW/m^2K) and bulk-like thermal conductivity for the $LaAlO_3$ film (13 W/m K)[27] [black dashed line, Fig. 4]; (2) high interfacial thermal conductance (350 MW/m^2K) and diminished thermal conductivity for the $LaAlO_3$ film (6 W/m K)[27] [blue dashed line, Fig. 4]; and (3) diminished interfacial thermal conductance (250 MW/m^2K) and diminished thermal conductivity for the $LaAlO_3$ film (6 W/mK) [red dashed line, Fig. 4]. Values for the heat capacity, thermal resistance across the Al/$LaAlO_3$ interface (150 MW/m-K), and the thermal conductivity of non-stoichiometric $LaAlO_3$ films are based on the experimentally measured values.[27]

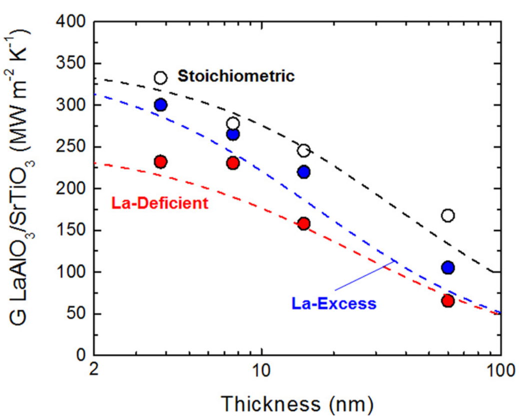

FIG. 4. Effective thermal conductance, G, for $LaAlO_3/SrTiO_3(001)$ heterostructures probed as a function of film thickness for films series controlled to possess 4%–5% La-deficient (red data), nearly stoichiometric (black data), and 4%–5% La-excess (blue data) film stoichiometry. The G for films <10 nm thick are dominated by the thermal properties of the interface, while for those on 60 nm thick films probe, the thermal properties of the film as well. Three hypothetical models (dashed lines) are provided for context: (1) a film with a high interfacial thermal conductance (here assumed to be 350 MW/m^2K) and bulk-like thermal conductivity for the $LaAlO_3$ film (13 W/mK) (black dashed line); (2) a film with a high interfacial thermal conductance (350 MW/m^2K) and diminished thermal conductivity for the $LaAlO_3$ film (6 W/mK) (blue dashed line); and (3) a film with a diminished interfacial thermal conductance (250 MW/m^2K) and diminished thermal conductivity for the $LaAlO_3$ film (6 W/mK) (red dashed line).

In first exploring the La-excess films, we observe a high G-value ($\sim$300 MW/m^2K), consistent with what we would expect for a system with a high-quality interface. As the film thickness is increased, the effective thermal conductance begins to decrease, roughly following the dashed blue line. Thus, we can understand the La-excess films to possess a high-quality interface, but a relative low bulk thermal conductivity (consistent with prior studies of thermal conductivity in (non-)stoichiometric $LaAlO_3$ films, where divergence from ideal cation stoichiometry reduces the thermal conductivity).[27] For nearly stoichiometric films, we once again note a high conductance value for the thinnest films (>325 MW/m^2K) and an overall decrease in effective thermal conductance with increasing film thickness—consistent with the model for a film possessing high interfacial thermal conductance and bulk-like thermal conductivity for the $LaAlO_3$ film (again consistent with prior studies of thermal conductivity in stoichiometric $LaAlO_3$ thin films, which possess the highest thermal conductivities).[27] This indicates that nearly stoichiometric films possess both a high-quality interface and high thermal conductance in the bulk of the film. Finally, for the La-deficient films, we observe relatively low effective thermal conductance values for both the thin and thick films and a good match to the model for a film with a diminished interfacial thermal conductance and diminished thermal conductivity for the $LaAlO_3$ film. While prior studies have already established that La-deficient films possess reduced thermal conductivity as compared to stoichiometric and bulk $LaAlO_3$,[27] this study indicates that the interfaces of the La-deficient $LaAlO_3/SrTiO_3$ heterostructures are also significantly more defective than for the stoichiometric and La-excess films. Therefore, consistent with our electrical transport data, our thermal studies indicate that La-excess and stoichiometric films possess a near-ideal interface, while La-deficient films possess a highly defective (reduced) near-interface portion of the $SrTiO_3$.

Ultimately, there is a growing set of evidence which suggests a non-trivial chemical component to the evolution of interfacial conductivity in $LaAlO_3/SrTiO_3$. The thermal measurements suggest that while both La-excess and La-deficient films possess diminished bulk thermal conductivity, only the La-deficient films also exhibit a greatly diminished interfacial thermal conductance. This likely arises from the added kinetic energy of the growth process that produces La-deficient films and the (non)stoichiometry itself, which drives disorder and reduction within the $SrTiO_3$ (thereby inducing enhanced conductivity).

To summarize, we have explored the combined effects of film stoichiometry, thickness, and oxygen pressure during growth in influencing the electrical and thermal transport properties of $LaAlO_3/SrTiO_3$ heterointerfaces. The electrical resistivity of this heterointerface can be tuned using laser fluence, all films (grown at sufficiently oxidizing conditions) exhibit a critical thickness of 3–4 uc for the onset of conduction, and that the nature of the conductance is highly influenced by the stoichiometry of the $LaAlO_3$ with La-deficient samples showing dramatic changes with thickness. TDTR studies show a diminished interfacial thermal conductance for the La-deficient films when compared to La-excess and stoichiometric films, suggesting that the interfacial conductance is more influenced by extrinsic factors (defects, disorder) at and near the interface. In turn, we suggest that although electronic reconstruction driven by a polar catastrophe appears to be present and to play a role in the conduction, the growth process and resulting film can push the system towards 3-dimensional conduction. Ultimately, controlling these materials requires a key understanding of the materials chemistry and the ability to control these factors on a fine scale.

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[1]A. Ohtomo and H. Y. Hwang, Nature **427**, 423 (2004).
[2]Y.-W. Xie and H. Y. Hwang, Chin. Phys. B **22**, 127301 (2013).
[3]A. Brinkman, M. Huijben, M. van Zalk, J. Juijben, U. Zeitler, J. C. Maan, W. G. van der Wiel, G. Rijnders, D. H. A. Blank, and H. Hilgenkamp, Nat. Mater. **6**, 493 (2007).
[4]N. Reyren, S. Thiel, A. D. Caviglia, L. Fitting-Kourkoutis, G. Hammerl, C. Richter, C. W. Schneider, T. Kopp, A.-S. Ruetschi, D. Jaccard, M. Gabay, D. A. Muller, J.-M. Triscone, and J. Mannhart, Science **317**, 1196 (2007).
[5]G. Singh-Bhalla, C. Bell, J. Ravichandran, W. Siemons, Y. Hikita, S. Salahuddin, A. F. Hebard, H. Y. Hwang, and R. Ramesh, Nat. Phys. **7**, 80 (2011).
[6]J. W. Park, D. F. Bogorin, C. Cen, D. A. Felker, Y. Zhang, C. T. Nelson, C. W. Bark, C. M. Folkman, X. Q. Pan, M. S. Rzchowski, J. Levy, and C.-B. Eom, Nat. Commun. **1**, 94 (2010).
[7]C. Cen, S. Thiel, G. Hammerl, C. W. Schneider, K. E. Anderson, C. S. Hellberg, J. Mannhart, and J. Levy, Nat. Mater. **7**, 298 (2008).
[8]P. Zubko, S. Gariglio, M. Gabay, P. Ghosez, and J. Triscone, Annu. Rev. Condens. Mater.Phys. **2**, 141 (2011).
[9]N. Nakagawa, H. Hwang, and D. Muller, Nat. Mater. **5**, 204 (2006).
[10]R. Pentcheva and W. E. Pickett, J. Phys.: Condens. Matter **22**, 043001 (2010).
[11]M. Huijben, A. Brinkman, G. Koster, G. Rijnders, H. Hilgenkamp, and D. H. A. Blank, Adv. Mater. **21**, 1665 (2009).
[12]J. N. Eckstein, Nat. Mater **6**, 473 (2007).
[13]A. Kalabukhov, R. Gunnarsson, J. Borjesson, E. Olsson, T. Claeson, and D. Winkler, Phys. Rev. B **75**, 121404 (2007).
[14]K. Shibuya, T. Ohnishi, M. Lippmaa, and M. Oshima, Appl. Phys. Lett. **91**, 232106 (2007).
[15]S. A. Chambers, M. H. Engelhard, V. Shutthanandan, Z. Zhu, T. C. Droubay, L. Qiao, P. V. Sushko, T. Feng, H. D. Lee, T. Gustafsson, E. Garfunkel, A. B. Shah, J. M. Zuo, and Q. M. Ramasse, Surf. Sci. Rep. **65**, 317 (2010).
[16]S. Thiel, G. Hammerl, A. Schmehl, C. W. Schneider, and J. Mannhart, Science **313**, 1942 (2006).
[17]P. R. Willmott, S. A. Pauli, R. Herger, C. M. Schlepuetz, D. Martoccia, B. D. Patterson, B. Delley, R. Clarke, D. Kumah, C. Cionca, and Y. Yacoby, Phys. Rev. Lett. **99**, 155502 (2007).
[18]F. Schoofs, T. Fix, A. S. Kalabukhov, D. Winkler, Y. Boikov, I. Serenkov, V. Sakharov, T. Claeson, J. L. MacManus-Driscoll, and M. G. Blamire, J. Phys.: Condens. Matter **23**, 305002 (2011).
[19]M. Golalikhani, Q. Y. Lei, G. Chen, J. E. Spanier, H. Ghassemi, C. L. Johnson, M. L. Taheri, and X. X. Xi, J. Appl. Phys. **114**, 027008 (2013).
[20]M. L. Reinle-Schmitt, C. Cancellieri, D. Li, D. Fontaine, M. Medarde, E. Pomjakushina, C. W. Schneider, S. Gariglio, P. Ghosez, J.-M. Triscone, and P. R. Willmott, Nat. Commun. **3**, 932 (2012).
[21]E. Breckenfeld, N. Bronn, J. Karthik, A. R. Damodaran, S. Lee, N. Mason, and L. W. Martin, Phys. Rev. Lett. **110**, 196804 (2013).
[22]H. K. Sato, C. Bell, Y. Hikita, and H. Y. Hwang, Appl. Phys. Lett. **102**, 251602 (2013).
[23]M. P. Warusawithana, C. Richter, J. A. Mundy, P. Roy, J. Ludwig, S. Paetel, T. Heeg, A. A. Pawlicki, L. F. Kourkoutis, M. Zheng, M. Lee, B. Mulcahy, W. Zander, Y. Zhu, J. Schubert, J. N. Eckstein, D. A. Muller, C. Stephen Hellberg, J. Mannhart, and D. G. Schlom, Nat. Commun. **4**, 2351 (2013).
[24]M. Kawasaki, K. Takahashi, T. Maeda, R. Tsuchiya, M. Shinohara, O. Ishiyama, T. Yonezawa, M. H. Yoshimoto, and H. Koinuma, Science **266**, 1540 (1994).
[25]T. Ohnishi, K. Shibuya, M. Lippmaa, D. Kobayashi, H. Kumigashira, and M. Oshima, Appl. Phys. Lett. **85**, 272 (2004).
[26]E. Breckenfeld, R. Wilson, J. Karthik, A. R. Damodaran, D. G. Cahill, and L. W. Martin, Chem. Mater. **24**, 331 (2012).
[27]E. Breckenfeld, R. B. Wilson, and L. W. Martin, Appl. Phys. Lett. **103**, 082901 (2013).
[28]J. Delahaye and T. Grenet, J. Phys. D **45**, 315301 (2012).
[29]Y. Z. Chen, N. Bovet, F. Trier, D. V. Christensen, F. M. Qu, N. H. Andersen, T. Kasama, W. Zhang, R. Giraud, J. Dufouleur, T. S. Jesperson, J. R. Sun, A. Smith, J. Nygard, L. Lu, B. Buchner, B. G. Shen, S. Linderoth, and N. Pryds, Nat. Commun. **4**, 1371 (2013).
[30]X. Luo, B. Wang, and Y. Zheng, Phys. Rev. B **80**, 104115 (2009).
[31]D. G. Cahill, W. K. Ford, K. E. Goodson, D. G. Mahan, A. Majumdar, H. J. Maris, R. Merlin, and S. R. Phillpot, J. Appl. Phys. **93**, 793 (2003).
[32]D. G. Cahill, Rev. Sci. Instrum. **75**, 5119 (2004).
[33]W. S. Capinski, H. J. Maris, T. Ruf, M. Cardona, K. Ploog, and D. S. Katzer, Phys. Rev. B **59**, 8105 (1999).
[34]See supplementary material at http://dx.doi.org/10.1063/1.4896778 for a complete description of the sensitivity of thermal models to the various parameters of interest.
[35]S.-M. Lee and D. G. Cahill, Microscale Thermophys. Eng. **1**, 47 (1997).