Confinement-driven metal-insulator transition and polarity-controlled conductivity of epitaxial LaNiO₃/LaAlO₃ (111) superlattices

Haoming Wei, Marius Grundmann, and Michael Lorenz

Citation: *Appl. Phys. Lett.* **109**, 082108 (2016); doi: 10.1063/1.4961693

View online: <https://doi.org/10.1063/1.4961693>

View Table of Contents: <http://aip.scitation.org/toc/apl/109/8>

Published by the American Institute of Physics

Articles you may be interested in

[Epitaxial growth of \(111\)-oriented LaAlO₃/LaNiO₃ ultra-thin superlattices](#)

Applied Physics Letters **101**, 261602 (2012); 10.1063/1.4773375

[Charge transfer-induced magnetic exchange bias and electron localization in \(111\)- and \(001\)-oriented LaNiO₃/LaMnO₃ superlattices](#)

Applied Physics Letters **110**, 102403 (2017); 10.1063/1.4978358

[Modeling the conductivity around the dimensionality-controlled metal-insulator transition in LaNiO₃/LaAlO₃ \(100\) superlattices](#)

Applied Physics Letters **106**, 042103 (2015); 10.1063/1.4907011

[Low-dimensional Mott material: Transport in ultrathin epitaxial LaNiO₃ films](#)

Applied Physics Letters **96**, 062114 (2010); 10.1063/1.3309713

[Electronic structure of buried LaNiO₃ layers in \(111\)-oriented LaNiO₃/LaMnO₃ superlattices probed by soft x-ray ARPES](#)

APL Materials **5**, 016101 (2017); 10.1063/1.4973558

[Comparative study of LaNiO₃/LaAlO₃ heterostructures grown by pulsed laser deposition and oxide molecular beam epitaxy](#)

Applied Physics Letters **110**, 041606 (2017); 10.1063/1.4975005

AIP | Conference Proceedings

**Get 30% off all
print proceedings!**

Enter Promotion Code **PDF30** at checkout

Confinement-driven metal-insulator transition and polarity-controlled conductivity of epitaxial $\text{LaNiO}_3/\text{LaAlO}_3$ (111) superlattices

Haoming Wei, Marius Grundmann, and Michael Lorenz^{a)}

Semiconductor Physics Group, Institut für Experimentelle Physik II, Universität Leipzig, Linnéstr. 5, 04103 Leipzig, Germany

(Received 30 June 2016; accepted 15 August 2016; published online 26 August 2016)

Recently, topological conductivity has been predicted theoretically in LaNiO_3 (111)-based superlattices. Here we report high-quality epitaxial $\text{LaNiO}_3/\text{LaAlO}_3$ superlattices on (111)-oriented SrTiO_3 and LaAlO_3 single crystals. For both substrates a metal-insulator transition with decreasing number of LaNiO_3 monolayers is found. While the electrical transport is dominated by two-dimensional variable range hopping for superlattices grown on polar mismatched SrTiO_3 (111), it switches to a thermally activated single gap behavior on polar matched LaAlO_3 (111). The gap energy of the polar double-layer LaNiO_3 superlattices can be tuned via the thickness of the insulating LaAlO_3 layers. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4961693>]

Controlling the band structure of transition metal oxides by designing artificial superlattices (SLs) has provided a way to explore quantum states at the interfaces. Particularly interesting in this respect are LaNiO_3 (LNO)-based superlattices. LaNiO_3 (LNO) is an interesting perovskite oxide crystallizing in a three-dimensional nearly cubic structure with a small rhombohedral distortion ($\beta = 90.41^\circ$).^{1,2} LNO in bulk and thin film form has been extensively studied in the past, showing metallic paramagnetism, metal-insulator transition, and polar metallic behavior.^{3–5} Recently, LNO-based superlattices (SLs) have attracted significant attention as they may exhibit interface-related novel properties at interfaces that are not present in either of the constituent materials. For example, high-temperature superconductivity has been predicted due to the similarity of the electronic structures of $\text{LaNiO}_3/\text{LaAlO}_3$ (LNO/LAO) SLs and cuprate-like superconductors.^{6,7} More interesting, in so called (111)-oriented bilayer LNO SLs, i.e., two LNO monolayers are sandwiched between band insulator layers of LAO, the Ni ions form a buckled honeycomb lattice which gives rise to a Dirac point in the Brillouin zone.^{8–10} As predicted by density functional theory calculations, an opening of the energy gap at these Dirac points combined with proper filling of the resulting states may result in a Z_2 topologically conducting behavior.^{11,12}

Experimental work is so far mostly focused to (100)-oriented LNO/LAO SLs which can be grown with high precision on different substrates. Detemple *et al.* grew LNO/LAO SLs on (100) $(\text{La,Sr})\text{AlO}_4$ substrates with different single-layer thickness. Two types of Ruddlesden–Popper faults were found and their origin and exact atomic arrangement were discussed in detail.¹³ Liu *et al.* studied the polar discontinuity of LNO/LAO SLs grown on nonpolar (100) TiO_2 -terminated SrTiO_3 (STO) substrates.¹⁴ Quantum confinement effects on the electronic properties of SLs were investigated by *ab initio* cluster calculations together with absorption spectra.¹⁵ Boris *et al.* studied the electronic phase transitions in nickel-oxide SLs by optical ellipsometry and low-energy

muon spin rotation.¹⁶ Frano *et al.* presented an X-ray diffraction (XRD) and electron microscopy study which indicated the overall thickness of an SL serving as a control parameter for the lattice symmetry of its individual components.¹⁷ The lattice distortion and octahedral rotation of epitaxially strained SLs on $(\text{La,Sr})\text{AlO}_4$ (100) were studied by Kinyanjui *et al.*¹⁸ More recently, we reported a dimensionality-controlled metal-insulator transition in (100) LNO/LAO SLs and the corresponding conductivity mechanisms were discussed by means of appropriate modeling.¹⁹

In contrast, very little experimental effort has been directed to (111)-oriented substrates due to the highly polar atomic layers along the [111] direction. These polar planes have a significant effect on the growth and may lead to a surface reconstruction.²⁰ In 2012, Middey *et al.* reported the synthesis of LNO/LAO heterostructures with 2/*m* monolayers, respectively, on mixed-terminated LAO(111) substrates.²¹ Instead of the predicted Dirac-point semimetallic behavior, an activated transport was observed.

Motivated by the above mentioned, promising theoretical predictions,^{11,12} together with recent experimental progress in epitaxial growth of LNO-based films and SLs,^{19,22} we present here (111) $[\text{LNO}_m/\text{LAO}_n]_l$ SLs (*m* and *n* indicate the number of unit cells, respectively, thereafter referred to as $[\text{m}/\text{n}]_l$). The stacking periodicity is repeated *l* times to adjust the total superlattice thickness. The SLs were deposited using pulsed laser deposition (PLD) with a KrF excimer laser; see Ref. 23 and references therein. The SL structure is schematically illustrated in the inset of Fig. 1(b). Two sintered stoichiometric LNO and LaAlO_3 (LAO) pellets were used as PLD targets. As substrates, (111)-oriented LAO, SrTiO_3 (STO) and Nb-doped STO single crystals with selected miscut were used. Both LAO(111) and STO(111) are strongly polar and consist of alternating $\text{LaO}_3^{3-}\text{Al}^{3+}$ or $\text{SrO}_3^{4-}\text{Ti}^{4+}$ charged planes, respectively, which are stacked along the [111] direction. The PLD growth was performed at temperatures of about 680 °C at an oxygen pressure of 0.05 mbar. After deposition, samples were annealed *in-situ* in 800 mbar oxygen at 680 °C for 15 min. A DC sputter coater with a designed shadow mask was applied to deposit

^{a)} Author to whom correspondence should be addressed. Electronic mail: mlorenz@physik.uni-leipzig.de

FIG. 1. (a) XRD 2θ - ω -scans of PLD grown (111) oriented LNO films on STO (111). Inset shows typical XRD φ -scans of the LNO (110) and STO (110) reflexes. (b) XRD 2θ - ω -scans of a $[9/7]_{10}$ SL on STO (111) substrate. The inset shows a scheme of the SL. (c) RSM of $[9/7]_{10}$ SL around the asymmetric STO (321) peak measured with $K\alpha_{1/2}$ radiation. (d) AFM image of $[2/2]_6$ SL on Nb-doped STO (111). The line scan shows the terrace steps heights of about 0.217 nm. Straight red lines are added as guide to the eyes.

80 nm thin gold contacts as shown in Fig. S1 in the [supplementary material](#).

The crystalline structure of the samples was analyzed by XRD with Cu-K α radiation and a Bragg-Brentano goniometer with focusing beam optics and secondary graphite monochromator to ensure low background signal. For the XRD φ -scan, the high-resolution four-circle goniometer of the Philips X'Pert diffractometer was used. Reciprocal space maps (RSM) were recorded with a PANalytical X'pert PRO MRD using Cu-K α radiation from a parabolic mirror and a PIXcel^{3D} multichannel detector. The interface and surface roughness, and layer thickness were investigated by X-ray reflectivity (XRR) and atomic force microscopy (AFM). Scanning transmission electron microscopy (STEM) and energy dispersive X-ray (EDX) mappings taken with a field emission scanning electron microscope FEI NOVA Nanolab 200 additionally confirm the periodic layer structure of the SLs. The in-plane resistivity was measured as a function of temperature in Van der Pauw geometry (Fig. S1) using a Hall setup.

Figure 1(a) shows XRD 2θ - ω -scan of the LNO film with a thickness of about 400 nm deposited on STO (111) substrate. The high (111) peak of LNO indicates the proper out-of-plane orientation. Not any impurity phases were detected. The broadening of LNO peak with a small shoulder peak indicates strain relaxation in this particularly thick LNO film. The φ -scan was carried out to investigate the in-plane epitaxial relationship of the LNO film to the STO (111) substrate. Three equally spaced peaks separated by 120° can be observed in the inset of Fig. 1(a). A threefold symmetry axis is clearly evident, corresponding to the rhombohedral and cubic structure of LNO film and STO substrate, respectively. This confirms the epitaxial relations LNO [111] || STO [111] and LNO [110] || STO [110] for out-of-plane and in-plane direction, respectively.

Figure 1(b) displays the XRD 2θ - ω -scan of a $[9/7]_{10}$ SL on STO (111). The SL Bragg peak is visible near STO (111).

substrate peak. The average distance between (111) layers of the SL was calculated to be around 0.21 nm from the position of SL (111) peak. Fig. 1(c) shows a representative XRD RSM around the (321) reflection of STO (111) substrate. The in-plane lattice match of [9/7]₁₀ SL and STO (111) substrate, i.e., the strained, pseudomorphic growth is visually confirmed by the vertical alignment of the (321) substrate and SL peaks at identical $q_{||}$ values. Fig. 1(d) shows the smooth monolayer structure of the surface of the SLs with root mean square roughness of 0.166 nm. The line scan shows the average step height of about 0.217 nm corresponding to the above mentioned value obtained by XRD.

For a direct impression of the SL periodicity, Fig. 2(a) depicts the STEM and EDX mapping results of a thicker [94/96]₁₀ SL. The STEM cross section and the corresponding EDX map of Ni confirm the presence of 10 double layers LNO/LAO with reasonably smooth interfaces. XRR, shown in Fig. 2(b), was used to determine the thickness and interfacial roughness of the thinner SLs. The strong oscillations arising from the finite thickness of the layers are clearly resolved for all SLs, indicating excellent sample quality with abrupt interfaces. Taking the [9/7]₁₀ SL, for example, by fitting the XRR curve, the average thickness of each LNO and LAO single layer is 1.91 ± 0.01 nm and 1.51 ± 0.01 nm, respectively, in agreement with our design of 9 LNO and 7 LAO unit cells. The interfacial roughness is in between 0.56 and 0.58 nm.

In our SLs, we observe a clear dependence of the sheet resistance on the number of LNO unit cells in SLs. As shown in Fig. 3(a), each insulating layer is formed by 7 LAO unit cells. The thick insulating layers ensure that the electronic transport near the Fermi energy is effectively confined to the LNO layers. The SLs were deposited on STO and LAO substrates which induce tensile (1.74%) and compressive (-1.27%) strain, respectively. The $[18/7]_{10}$ SL shows metallic

FIG. 2. (a) STEM dark-field image of $[94/96]_{10}$ SL grown on STO (111) substrate. The EDX map (inset) shows the distribution of Ni in the SL. (b) X-ray reflectivity of LNO/LAO SLs. The notation refers to the structure of SLs on the indicated substrate materials.

behavior with positive resistivity temperature coefficient at temperature range from 70 K to 300 K. With decreasing number of LNO unit cells in the SLs, insulating behavior appears. Obviously, the metal-insulator transition is related to quantum confinement when lowering the dimensionality of the LNO layers in the SLs which has been discussed previously for the (100) orientation.¹⁹

The $[7/6]_{10}$ and $[7/4]_{10}$ SLs on STO are highly insulating hinting to a possible Mott behavior as shown in Fig. 3(b). The logarithm of conductance is proportional to $T^{-1/3}$, i.e., it can be described with a two-dimensional (2D) Mott variable range hopping (VRH) model, $\sigma = \sigma_0 \exp[-(T_0/T)^{1/3}]$.²⁴ T_0 is the localization temperature that depends on the density of states $N(E_F)$ near the Fermi level. T_0 can be obtained by fitting the conductance curve. For the validity of the model, two criteria have to be fulfilled:²⁵ $R_{hop}/a > 1$ and $E_{hop}/k_B T > 1$, where R_{hop} is the mean hopping distance, a is the localization length, and E_{hop} is the mean hopping energy. According to the Mott theory of 2D VRH, their relationships are given as: $R_{hop} = \frac{1}{3}a(\frac{T_0}{T})^{1/3}$ (Ref. 26) and $E_{hop} = \frac{1}{3}k_B T^{2/3} T_0^{1/3}$ (Ref. 27). For our samples, the ratios R_{hop}/a and $E_{hop}/k_B T$ derived from the fitting satisfy the criteria within the measured temperature range and thus confirm the validity of the 2D VRH model.

FIG. 3. (a) Temperature-dependent sheet resistance of (111) SLs deposited on STO (red) and LAO (blue) substrates. The horizontal black dotted line corresponds to $25.8 \text{ k}\Omega/\square$, the quantum of resistance in 2D state. (b) Logarithm of conductance $\ln(\sigma)$ as a function of $T^{-1/3}$ of $[6/7]_{10}$ and $[4/7]_{10}$ SLs on STO substrate, indicating 2D VRH-type conductivity. For comparison, the corresponding data of the $[4/7]_{10}$ SL on LAO are shown in Fig. S2 in the [supplementary material](#).

It is also possible to estimate the value of localization length a by using the relationship:²⁸ $T_0 = 13.8/k_B N(E_F) a^2$. Assuming that the Fermi energy lies in the range of the localized states and $N(E_F)$ values are on the same order of magnitude within several $k_B T$, we can take $k_B N(E_F) = \partial n / \partial T \approx n(100 \text{ K}) / 100 \text{ K}$ as a rough approximation.²⁹ The calculated localization length a of our SLs are 1.2 and $2.3 \text{ \AA}$, which is reasonable in comparison with the Ni-O bond distance in SLs on STO of about $2 \text{ \AA}$. All fitting parameters and calculated values are listed in Table I.

Compared with SLs deposited on LAO, the SLs on STO have a higher sheet resistance for all structures as shown in Fig. 3(a). A possible reason is the substrate-induced strain in the in-plane lattice-matched SLs which may influence the value of resistivity. Hence, a possible reason for the enhancement of resistivity of SLs on STO could be the slight expansion of their in-plane lattice constants, which leads to larger Ni-O-Ni bond lengths.²²

However, the transport behavior of SLs on LAO substrate cannot be described by the 2D Mott VRH model because the calculated hopping distance is much smaller than the localization length. The criterion $R_{hop}/a > 1$ is not fulfilled for $[6/7]_{10}$ SL on LAO which undergoes a metal-insulator transition with the sheet resistance around $25.8 \text{ k}\Omega/\square$.¹⁹ In addition, a comparison of sheet conductance of $[4/7]_{10}$ SL on LAO as a function of $T^{-1/3}$ and $1000/T$ is shown in Fig. S2 in the [supplementary material](#). The different temperature dependence of resistivity for SLs on STO and LAO may be due to the substrate polarity and indicates changing conductivity mechanism which will be discussed in the following. As mentioned above, the STO

TABLE I. Fitting parameters and calculated values of the 2D VRH model for two selected SLs on STO (111): Sheet resistance R_S at 300 K, density of states at the Fermi level $N(E_F)$, fit parameter T_0 , localization length a , the ratio of R_{hop}/a at 300 K, and hopping energy E_{hop} .

SLs	R_S (T = 300 K) ($\Omega/\square$)	$N(E_F)$ ($\text{eV}^{-1}\text{cm}^{-2}$)	T_0 (K)	a ($\text{\AA}$)	R_{hop}/a (T = 300 K)	E_{hop} (meV)
[6/7] ₁₀	1.36×10^4	6.14×10^{20}	1.83×10^4	1.19	1.31	$0.76 T^{2/3}$
[4/7] ₁₀	1.34×10^5	1.42×10^{19}	2.06×10^5	2.33	2.94	$1.70 T^{2/3}$

substrates consist of alternating $+4e$ and $-4e$ charged atomic planes along the [111] direction which leads to a large polarity mismatch at the interface to the LNO/LAO SLs. In contrast, the interface is polar matched for SLs grown on LAO substrate with sequentially $+3e$ and $-3e$ charged atomic planes along [111]. Different polarity of the substrates may induce the changing of Ni valance state which has been observed in LNO/LAO (100) SLs.¹⁴ Since the conductivity of LNO strongly depends on the valence states of Ni, we attribute the different conductivity mechanisms mainly to the substrate-induced polarity.

Figure 4(a) shows the evolution of sheet resistance with temperature for the [2/3]₁₀ SL on LAO. The resistivity data are first analysed using the Mott-VRH model which can describe the transport of LNO thin films and (100)-oriented SLs well.^{4,19} However, the fitting quality is really poor for (111)-oriented SLs on LAO in the high temperature range (not shown). Instead, an activated transport model fits the data in the temperature range from 110 K to 300 K.²¹ The characteristic relation is: $\sigma \propto \exp(-E_g/2k_B T)$, where E_g

denotes the band gap. As shown in the inset of Fig. 4(a), linear fitting of [2/3]₁₀ SL yields a value of E_g of about 107 meV which is close to another reported value of about 95 meV obtained by the same model.²¹ A possible origin of the gap structure is the breaking of the Z_2 symmetry of the two Ni sites in the bilayer LNO SLs.¹² As predicted by theoretical work, two triangular NiO_6 octahedron layers form a buckled honeycomb lattice which gives rise to Dirac points in the band structure.¹¹ Allowing full lattice relaxation breaks the inversion symmetry Z_2 and results in two inequivalent Ni sites which destroys the Dirac point by opening a gap at K and K' .¹² Below 90 K, the transport of SLs on LAO can be described using the 2D VRH model which has been discussed above.

To obtain further insight into the transport properties of bilayer LNO SLs, we changed the thickness of the LAO layers. As shown in Fig. 4(b), activated transport dominates in all bilayer SLs in the measured temperature range from 110 K to 300 K. With increasing the LAO thickness, the sheet resistance and activated gap values increase as listed in Table II. The tendency of developing a larger gap together with higher resistivity may be due to the accumulation of disorder which is expected for polar layer growth on the (111)-oriented substrate.²⁰ In addition, the insulating layers in the three [2/2] SLs are very thin. With increasing stacking periodicity, electrical conduction through insulating layers may occur which leads to a higher total conductance and smaller calculated band gap. Angle resolved photoemission spectroscopy (ARPES) has been tried to get deeper insight into detailed band structure features of our SLs. However, the ultra-high vacuum conditions of 2×10^{-10} mbar in the ARPES chamber at the synchrotron BESSY II influenced the sensitive monolayer surface structure (Fig. 1(d)) of our as-grown oxide SLs which makes such measurements difficult. Thus, at this stage we cannot make final statements about the theoretically predicted Z_2 topological behavior of our SLs.¹²

In summary, a series of epitaxial LNO/LAO SLs has been grown on polar mismatched STO (111) and polar matched LAO (111) substrates. Strained, in-plane lattice

FIG. 4. (a) Temperature-dependent sheet resistances of [2/3]₁₀ SL deposited on LAO (111) substrate. The inset shows a linear fitting of sheet conductance between 300 and 110 K. (b) Sheet conductance of the bilayer LNO SLs as a function of $1000/T$. The notation refers to the structure of SLs. The fits confirm a thermally activated conductivity behavior on LAO (111).

TABLE II. Electrical characteristics of the bilayer LNO SLs on LAO (111) with increasing LAO layers thickness: sheet resistance R_S at 300 K and activated gap energy E_g .

SLs	R_S (T = 300 K) ($\Omega/\square$)	E_g (meV)
[2/2] ₈	1.29×10^4	17.3 ± 0.1
[2/2] ₁₀	7.78×10^3	10.2 ± 0.1
[2/2] ₁₂	6.59×10^3	5.9 ± 0.1
[2/3] ₁₀	3.18×10^5	107 ± 1
[2/4] ₁₀	3.77×10^5	120 ± 1
[2/5] ₁₀	1.41×10^6	154 ± 2
[2/6] ₁₀	1.70×10^6	162 ± 2

matched growth was shown by XRD analysis. Smooth 2-dimensional surface and interfaces are demonstrated by AFM, XRR, and STEM. A transition from metallic to VRH or thermally activated transport was observed in the (111) SLs when decreasing the LNO thickness on STO or LAO substrates, respectively. While the absolute resistivity values depend on the thickness-dependent strain, its temperature dependence, i.e., the resistivity mechanism, depends on the polar mismatch/match to the used substrate material. Instead of the predicted Dirac-point semimetals, thermally activated behavior appears in the bilayer LNO SLs on LAO (111) which should be, from theory, a potential candidate for a 2D topological insulator. The gap energy can be tuned by changing the thickness of LAO layers. The understanding of these transport properties is essential for the design and fabrication of artificial SLs with tailored electronic properties.

Supplementary Material shows first a scheme and a photograph of a typical superlattice sample with contacts for the in-plane resistivity measurements and second the conductivity data of the $[4/7]_{10}$ superlattice on LaAlO_3 for two different models, for direct comparison with Fig. 3(b).

We are grateful to the DFG for funding the Collaborative Research Center SFB 762 “Functionality of Oxide Interfaces.” We thank Oliver Rader from the Humboldt Center Berlin for the ARPES experiments, Jörg Lenzner for the STEM images, and Gabriele Ramm for PLD target preparation.

- ¹K. Tsubouchi, I. Ohkubo, H. Kumigashira, Y. Matsumoto, T. Ohnishi, M. Lippmaa, H. Koinuma, and M. Oshima, *Appl. Phys. Lett.* **92**, 262109 (2008).
- ²R. Eguchi, A. Chainani, M. Taguchi, M. Matsunami, Y. Ishida, K. Horiba, Y. Senba, H. Ohashi, and S. Shin, *Phys. Rev. B* **79**, 115122 (2009).
- ³J. Son, P. Moetakef, J. M. LeBeau, D. Ouellette, L. Balents, S. J. Allen, and S. Stemmer, *Appl. Phys. Lett.* **96**, 062114 (2010).
- ⁴R. Scherwitzl, S. Gariglio, M. Gabay, P. Zubko, M. Gibert, and J.-M. Triscone, *Phys. Rev. Lett.* **106**, 246403 (2011).
- ⁵T. H. Kim, D. Puggionoo, Y. Yuan, L. Xie, H. Zhou, N. Campbell, P. J. Ryan, Y. Choi, J. W. Kim, J. R. Patzner, S. Ryu, J. P. Podkaminer, J. Irwin, Y. Ma, C. J. Fennie, M. S. Rzchowski, X. Q. Pan, V. Gopalan, J. M. Rondinelli, and C. B. Eom, *Nature* **533**, 68–72 (2016).

- ⁶P. Hansmann, X. P. Yang, A. Toschi, G. Khaliullin, O. K. Andersen, and K. Held, *Phys. Rev. Lett.* **103**, 016401 (2009).
- ⁷P. Hansmann, A. Toschi, X. P. Yang, O. K. Andersen, and K. Held, *Phys. Rev. B* **82**, 235123 (2010).
- ⁸A. Rüegg and G. A. Fiete, *Phys. Rev. B* **84**, 201103(R) (2011).
- ⁹K. Y. Yang, W. G. Zhu, D. Xiao, S. Okamoto, Z. Q. Wang, and Y. Ran, *Phys. Rev. B* **84**, 201104(R) (2011).
- ¹⁰A. Rüegg, C. Mitra, A. A. Demkov, and G. A. Fiete, *Phys. Rev. B* **85**, 245131 (2012).
- ¹¹D. Xiao, W. Zhu, Y. Ran, N. Nagaosa, and S. Okamoto, *Nat. Commun.* **2**, 596 (2011).
- ¹²D. Doennig, W. E. Pickett, and R. Pentcheva, *Phys. Rev. B* **89**, 121110(R) (2014).
- ¹³E. Detemple, Q. M. Ramasse, W. Sigle, G. Cristiani, H. U. Habermeier, B. Keimer, and P. A. Aken, *J. Appl. Phys.* **112**, 013509 (2012).
- ¹⁴J. Liu, M. Kareev, S. Prosandeev, B. Gray, P. Ryan, J. W. Feeland, and J. Chakhalian, *Appl. Phys. Lett.* **96**, 133111 (2010).
- ¹⁵J. Liu, S. Okamoto, M. Veenendaal, M. Kareev, B. Gray, P. Ryan, J. W. Feeland, and J. Chakhalian, *Phys. Rev. B* **83**, 161102(R) (2011).
- ¹⁶A. V. Boris, Y. Matiks, E. Benckiser, A. Frano, P. Popovich, V. Hinkov, P. Wochner, M. C. Colin, E. Detemple, V. K. Malik, C. Bernhard, T. Prokscha, A. Suter, Z. Salman, E. Morenzoni, G. Cristiani, H. U. Habermeier, and B. Keimer, *Science* **332**, 937–940 (2011).
- ¹⁷A. Frano, E. Benckiser, Y. Lu, M. Wu, M. Castro-Colin, M. Reehuis, A. V. Boris, E. Detemple, W. Sigle, P. Aken, G. Cristiani, G. Logvenov, H.-U. Habermeier, P. Wochner, B. Kermer, and V. Hinkov, *Adv. Mater.* **26**, 258–262 (2014).
- ¹⁸M. K. Kinyanjui, Y. Lu, N. Gauquelin, M. Wu, A. Frano, P. Wochner, M. Reehuis, G. Christiani, G. Logvenov, H.-U. Habermeier, G. A. Botton, U. Kaiser, B. Keimer, and E. Benckiser, *Appl. Phys. Lett.* **104**, 221909 (2014).
- ¹⁹H. M. Wei, M. Jenderka, M. Bonholzer, M. Grundmann, and M. Lorenz, *Appl. Phys. Lett.* **106**, 042103 (2015).
- ²⁰S. Middey, P. Rivero, D. Meyers, M. Kareev, X. Liu, Y. Cao, J. W. Feeland, S. Barraza-Lopez, and J. Chakhalian, *Sci. Rep.* **4**, 6819 (2014).
- ²¹S. Middey, D. Meyers, M. Kareev, E. J. Moon, B. A. Gray, X. Liu, J. W. Feeland, and J. Chakhalian, *Appl. Phys. Lett.* **101**, 261602 (2012).
- ²²H. M. Wei, M. Jenderka, M. Grundmann, and M. Lorenz, *Phys. Status Solidi A* **212**, 1925–1930 (2015).
- ²³M. Lorenz and R. Rao, *J. Phys. D: Appl. Phys.* **47**, 030301 (2014).
- ²⁴W. Brenig, *Philos. Mag.* **27**, 1093 (1973).
- ²⁵M. Kneiß, M. Jenderka, K. Brachwitz, M. Lorenz, and M. Grundmann, *Appl. Phys. Lett.* **105**, 062103 (2014).
- ²⁶Z. H. Khan, M. Husain, T. P. Perng, N. Salah, and S. Habib, *J. Phys.: Condens. Matter* **20**, 475207 (2008).
- ²⁷S. J. Lee and J. B. Ketterson, *Phys. Rev. B* **46**, 12695 (1992).
- ²⁸M. I. B. Utama, X. Lu, D. Zhan, S. T. Ha, Y. Yuan, Z. Shen, and Q. Xiong, *Nanoscale* **6**, 12376–12382 (2014).
- ²⁹G. Eda, C. Mattevi, H. Yamaguchi, H. Kim, and M. Chhowalla, *J. Phys. Chem. C* **113**, 15768 (2009).