Misfit Dislocation Guided Topographic and Conduction Patterning in Complex Oxide Epitaxial Thin Films

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Interfacial dissimilarity has emerged in recent years as the cornerstone of emergent interfacial phenomena, while enabling the control of electrical transport and magnetic behavior of complex oxide epitaxial films. As a step further toward the lateral miniaturization of functional nanostructures, this work uncovers the role of misfit dislocations in creating periodic surface strain patterns that can be efficiently used to control the spatial modulation of mass transport phenomena and bandwidth-dependent properties on a ≈20 nm length scale. The spontaneous formation of surface strain-relief patterns in La_{0.7}Sr_{0.3}MnO_3/LaAlO_3 films results in lateral periodic modulations of the surface chemical potential and tetragonal distortion, controlling the spatial distribution of preferential nucleation sites and the bandwidth of the epilayer, respectively. These results provide insights into the spontaneous formation of strain-driven ordered surface patterns, topographic and functional, during the growth of complex oxide heterostructures on lengths scales far below the limits achievable through top-down approaches.

1. Introduction

Thin film heteroepitaxy of complex oxides has evolved in recent years to a fascinating platform for the manipulation of materials properties by altering the subtle energy landscape of competing interactions through epitaxial strain and dissimilarity. Notably, this strategy has led to the discovery of exotic interfacial phenomena, while opening the possibility to tune the bulk transport, magnetic, ferroelectric, and multiferroic properties of thin films. However, next generation nanodevices demand a further step toward miniaturization, facing challenging strategies to controllably manipulate the lateral modulation of atomic length scales. In semiconductor epitaxy, this goal can be achieved through the Stranski–Krastanov growth mode, leading to the formation of self-assembled quantum dots on a wetting layer driven by lateral gradients in the surface chemical potential. This strategy, however, typically leads to nanostructures exhibiting broad size distributions and poor positional order. Strained films, on the other hand, usually relax by misfit dislocations (MDs) above a critical thickness at which their elastic energy exceeds the energy of the interfacial dislocation network. The overlapping of strain fields emanating from individual dislocations causes lateral modulations of lattice distortions which may extent up to the free surface. Therefore, highly organized MD networks buried at the substrate-film interface not only modulate the physical properties of thin films, but in addition can promote the growth of ordered nanostructures on their surfaces. In this sense, MDs have been used to produce strain guided patterned surfaces in semiconductor and metal systems, and more recently to tune Dirac surface states in topological insulators. However, to date, the extension of this concept to oxide epitaxy remains elusive.

A unique property of dislocations, that make them highly appealing for creating new functional nanostructures, is their multiscale character. While being essentially linear defects, they store their elastic energy at comparatively large distances (several nanometers) from their sub-nanometer core. As a consequence, they can modify the properties of the host material in two different length scales. On the one hand, dislocation lines can be considered as a separate phase exhibiting its own physical behavior. A clear manifestation of their singularity, for instance, comes from the observation that oxygen deficient dislocations in SrTiO_3 exhibit bistable resistive switching.
or that their nonsuperconducting cores induce pinning of the magnetic flux lines in type-II superconductors.\cite{24,25} On the other hand, owing to the strong sensitivity of the electronic structure to strain, MD long range strain fields are key for the realization of periodic functional (magnetic, electronic or catalytic)\cite{28} bulk and surface patterns. Particularly, in manganite thin films, theoretical studies show that the sign of the misfit strain profoundly alters their magnetotransport response.\cite{36} A compressive/tensile biaxial strain promotes a stronger/weaker Mn $3d$–$O 2p$ orbital overlapping leading to a broader/narrower bandwidth, allowing a local control on the electrical conduction by imposing spatial strain modulations. As MDs typically form square networks, they provide positional order along two mutually perpendicular directions for the creation of self-organized surface templates,\cite{13–17} in contrast with substrate steps which only induce unidirectional ordering.\cite{27} Notably, dislocation threading segments of the MDs induce a smearing of the ferroelectric phase transition of the image reveals a downward bending of atomic rows above the core. d) In-plane ($\varepsilon_{xx}$) and out-of-plane ($\varepsilon_{zz}$) strain maps, corresponding to the rectangular dashed window shown in panel (d). 

Figure 1. a) Cross-section HRTEM image of a 2 nm thick LSMO/LAO film, exhibiting a coherent, fully strained, interface (indicated by a horizontal arrow), and a flat surface. b) Cross-section HRTEM image corresponding to a 3.5 nm thick film, exhibiting a dislocated interface (an MD is shown inside the white circle) and an undulated surface (outgrowths are indicated by black arrowheads); the boxed area corresponds to the strain maps shown in panel (d). c) Magnified view of the dislocation core encircled in panel (b); white circles mark the corners of a Burgers circuit yielding Burgers vector, $\mathbf{b} = \theta_{LAO}[100]$; the two vertical arrows indicate two extra half-planes consistent with a split core consisting of two $1/2[100]$ partials; careful inspection of the image reveals a downward bending of atomic rows above the core. d) In-plane ($\varepsilon_{xx}$) and out-of-plane ($\varepsilon_{zz}$) strain maps, corresponding to the rectangular dashed window shown in panel (b), as determined from geometrical phase analysis (GPA) analysis; the two vertical arrows indicate the positions of the two partials building the split core; the surface of the film is indicated by a dashed line; the color scale bar corresponds to a full strain range of ±0.2 (negative and positive being compressive and tensile, respectively).

2. Results and Discussion

2.1. Dislocation Structure and Strain Evolution

Figure 1a shows a cross-section high resolution electron transmission microscopy (HRTEM) image of the 2 nm thick film, exhibiting coherent, dislocation free, interfaces and flat surfaces. The out-of-plane lattice parameter determined by high-resolution X-ray diffraction (HR-XRD) is 3.98(1) Å, which for a fully strained film yields a Poisson’s ratio $\nu = \varepsilon_{zz}/(\varepsilon_{zz} - 2\varepsilon_{xx}) = 0.35$, very
close to that derived for tensile strained LSMO/STO films in their elastic regime, \( \nu = 0.33 \). As shown in Figure 1b, the 3.5 nm thick film, on the other hand, exhibits an undulated surface and a dislocated interface (see the encircled MD). Careful inspection of this image shows that the surface undulations are due to both, a downward bending of the horizontal atomic rows above the dislocation core (see also Figure 1c), and outgrowths, as indicated by arrowheads, that as a general trend form on the surface at each side of the projected line of the buried MDs, \( \approx 4 \) nm away from them (see Figure 1b). In thicker films, the outgrowths are no longer present, while the bending of atomic planes prevails independently of film thickness, Figure S1 (Supporting Information).

An enlarged view of the core structure of the encircled MD is presented in Figure 1c. The Burgers circuit yields a Burgers vector \( \mathbf{b}_x = [100] \), parallel to the interface, as indicated in the figure. The two arrowed vertical atomic rows terminating at the slip plane bare witness of a split core: \([100] \rightarrow 1/2[100]+1/2[100] \) \((\mathbf{b}_x = \mathbf{b}_1 + \mathbf{b}_2)\). It is worthy to mention that similarly dissociated cores have been theoretically predicted for oxygen deficient \( \mathbf{b}_x = [100] \) edge dislocations in \( \text{SrTiO}_3 \),\(^{[22]}\) and also identified as a polymorphic form of \( \mathbf{b}_x = [100] \) dislocations in \( \text{MgO} \).\(^{[40]}\) In fact, analysis of several MD cores indicated that this core splitting is a general trend among the observed MDs. The two adjacent \( \mathbf{b}_1 \) and \( \mathbf{b}_2 \) partial dislocations build a rather compact core structure. The displacement field above the core is clearly manifested as a downward bending of the atomic rows, with a maximum amplitude of \( \approx 1.25 \) Å at the horizontal locus of the buried dislocation. Figure 1d shows maps of the in-plane, \( \varepsilon_{xx}(x,z) \), and out-of-plane, \( \varepsilon_{zz}(x,z) \), strain components around the MD, computed from the experimental image shown in panel (c) using Geometrical Phase Analysis.\(^{[41]}\) The positions of the two partials in the composite core are clearly discerned, along with the compressive (red, yellow) and tensile regions (magenta, blue) extending into the substrate and the film, respectively. It can be observed that the misfit relieving tensile strains, \( \varepsilon_{xx} \), draw two arms propagating up to the film surface, whilst normal strains, \( \varepsilon_{zz} \), rapidly vanish as a result of the boundary condition of a free (001) surface, \( \sigma_{xx} = \sigma_{zz} = \sigma_{yx} = 0 \) (\( \sigma_{ij} \) are components of the stress tensor).

Figure 2. a) AFM image showing the topography of a 3.5 nm thick film, consisting of ridges aligned with the [100] and [010] directions, and terraces; the color scale bar corresponds to a full range of 1.5 nm. b) Orientation contrast SEM images revealing the MD network of the same film. The scale bar is the same for both images. It can be observed that the ridge pattern mimics the underlying buried dislocation network.

A projection of the MD network on the interface plane obtained by orientation contrast scanning electron microscopy (SEM) is shown Figure 2a. The MD network is formed by irregularly spaced short \( \approx 100 \) nm dislocation segments along the [100] and [010] directions, with average spacing \( <S> = 25 \pm 10 \) nm (see Figure S1, Supporting Information). These lines correspond to the projection of half-loops onto the interfacial plane, where they leave a MD segment which increases in length as the threading segments move apart. With increasing film thickness to 6 nm, \( <S> \) reduces to \( 19 \pm 6 \) nm, while most of dislocations run across the limits of the imaged \( 1000 \) nm \( \times 1000 \) nm areas (see Figure S1, Supporting Information). The theoretical dislocation spacing for full relaxation, \( b_x/e = 16.5 \) nm, is achieved for the 14 nm thick film, with \( <S> = 16 \pm 3 \) nm. According to equilibrium theory, the energy barrier needed for a half-loop of critical radius to survive increases with decreasing misfit strain.\(^{[42]}\) Therefore, as the misfit is relieved by expansion of existing half-loops, the nucleation of new ones becomes kinetically suppressed. As a consequence, the average dislocation length increases whilst their lateral spacing narrows as the films thicken, in excellent agreement with experimental observations (see Figure S1, Supporting Information). Since the glide plane coincides with the interface plane, the MDs can easily rearrange their positions on that plane to minimize their elastic interactions, resulting in increasingly ordered patterns. According to this evolution, the density of threading arms of MDs rapidly decreases as the films thicken, while the strain state of the films becomes determined by the increasingly ordered interfacial MD network.

In order to analyze the morphological transition occurring between 2 and 3.5 nm, Figure 2 compares the MD network shown in panel (a) with a topographic atomic force microscopy (AFM) image of the same film panel (b). In agreement with the cross-section HRTEM image shown in Figure 1b, it is clearly seen that the surface topography consists of a network of ridges and terraces mimicking the underlying dislocation network. Hence, the outgrowths shown in Figure 1b correspond to a section of the ridges observed in the AFM image. To visualize the bulk strain state of the dislocated films, we obtained planar view strain sensitive low-angle annular dark field (LAADF)
images from the 6 nm thick film (see Figure 3a,b) and compared them with the spatial distribution of the residual in-plane strain ε_{xx}(x,y) + ε_0 (ε_0 = −2.3% is the background misfit strain), arising from a square network of b_x = [100] and b_y = [010] dislocations, using continuum isotropic elasticity (see Figure 3c). We employed the isotropic-average shear modulus of LSMO derived from the Voigt–Reuss–Hill averaging, \[ G = 50.98 \text{ GPa} \] using the cubic elastic stiffness coefficients reported by Darling et al. \[ G = 133.10 \text{ GPa} \]. Note that the LSMO is softer than the LAO substrate. In this situation, the core of the dislocation is predicted to lie in the film very close to the interface, in agreement with the present observations. The calculated strain map shows good match with the LAADF experimental image, in which darker areas correspond to a lower degree of misfit relaxation between dislocations. Minimum relaxation levels are also attained along the locus of dislocation lines and, to a lower extent, at the crossing points, as also resolved in the experimental image (see Figure 3b). This suggests that the observed topography results from preferential growth at surface sites with a minimum level of residual strain.

Since here we are interested on the effect of strains on surface phenomena, to confirm this hypothesis, here we estimate the strain state of the free film surface using displacement fields derived from the image dislocation approach. The vertical and horizontal atomic displacements, u_x and u_y, at the free surface of a film with thickness \( d \), induced by an interfacial dislocation with Burgers vector \( b_x \), are given by

\[ u_x(x) = \frac{b_x}{\pi} \left( \frac{d^2}{x^2 + d^2} \right) \]  
\[ u_y(x) = \frac{b_x}{\pi} \left[ \frac{-dx}{x^2 + d^2} + \tan^{-1}\left( \frac{x}{d} \right) \right] \]

From Equation (1), the amplitude of the downward atomic displacement at the dislocation coordinate \( x = 0 \) is \( b_x/\pi \approx 1.20 \text{ Å} \).
(b_x = a_{LSMO} = 3.79 Å), in excellent agreement with the displacement field determined from Figure 1c. The modulation of the surface strain is then obtained by differentiation of Equation (1b), \( \varepsilon_{xx}(x) = \partial u_M(x) / \partial x \)

\[
\varepsilon_{xx}(x) = \frac{b_d d}{\pi (x^2 + d^2) (x^2 + d^2 + 1)}
\]

(2)

The tensile strain given by Equation (2) is superimposed over the background compressive misfit strain due to the substrate, \( \varepsilon_0 \), yielding a residual surface strain, \( \varepsilon_{xx}(x) + \varepsilon_0 \), as plotted in Figure 4a for the 3.5 and 6 nm thick films, respectively. The figure includes three dislocations, the central one at \( x = 0 \). For the 3.5 nm thick film, assuming a dislocation spacing of 25 nm, almost total relaxation \( (\varepsilon_{xx}(x) + \varepsilon_0) = 0 \) occurs at each side of the dislocation line at a distance of about 4 nm. For the 6 nm thick one, assuming a dislocation spacing of 19 nm, the overlapping of strain fields erases the fluctuation between adjacent MDs. In this case complete relaxation is achieved within a region of size similar to the thickness of the film located between adjacent dislocations. However, for those two thicknesses, specially for the thinner film, the dispersion of \( S \) values is still high enough to expect the coexistence of both types of modulation in the same sample.

### 2.2. Evolution of Film Topography

The link between the morphological evolution of a free surface and its strain state is given by the surface chemical potential \( \mu(x) = \mu_0 + \gamma_2 \kappa(x) + \Omega w(x) \),\(^{[11]} \) where \( \mu_0 \) is the chemical potential of the unstrained flat surface, \( \gamma \) is the surface free energy per unit area, \( \Omega \) is the volume of a growth unit, \( \kappa(x) \) is the surface curvature, and \( w(x) \) is the local surface strain energy density.\(^{[48]} \) Assuming a flat surface, \( \kappa = 0 \), the modulation of the driving force for strain induced surface mass transport due to an underlying pure edge MD located at \( x = 0 \), \( \Delta \mu_{s}(x) = \mu(x) - \mu_0 = \Omega w(x) \), can be expressed as

\[
\Delta \mu_{s}(x) = \Omega M \left( \varepsilon_{xx}(x) + \frac{\varepsilon_{xx}^2(x)}{2 + 2w} \right)
\]

where \( M = 2G(1 + \nu)/(1 - \nu) \). According to Equation (3), growth units will migrate from highly strained regions to find stable positions at locations exhibiting minimum \( \Delta \mu_{s}(x) \) values at each side of the dislocation. To illustrate this, Figure 4b depicts the chemical potential reduction relative to the position of the MD, as a function of distance, \( x \): \( \Delta \mu_{s}(x) = \mu(x) - \mu_d \) (\( \mu_d \) is the surface chemical potential at the position of the MD) for the 3.5 and 6 nm thick films, with \( \langle S \rangle = 25 \) and 19 nm, respectively. For the 3.5 nm thick film, the fluctuation draws two minima at each side, \(-4 \) nm away from the MDs, where the chemical potential is reduced by 17 meV. This abrupt gradient in \( \Delta \mu_{s}(x) \) should induce the preferential nucleation of LSMO at each side of the MDs, leading to the formation of the observed topographic pattern. It should be noted, however, that since in this case the glide plane is parallel to the interface plane, MDs can easily move to rearrange their positions and, therefore, those surface features do not necessarily appear associated with them. It is to be noted that there exists a kinetic limitation for the vertical growth of the ridges: Once a ridge is formed, the rapid increase in surface curvature at that point causes the competition between the surface energy, \( \gamma \kappa(x) \), and strain energy, \( w(x) \), terms of the chemical potential, eventually hindering its vertical growth. This scenario drastically changes as the film surface moves further apart from the dislocation strain sources (and those ones get closer), as exemplified by the 6 nm thick film (see Figure 4b). The plot clearly shows that a thickness increase of 2.5 nm results in nearly a 50% decrease in the amplitude of the fluctuation in \( \Delta \mu_{s}(x) \). This effect contributes to homogenize the chemical potential throughout the surface, resulting in flatter films, in agreement with experimental observations (see Figure S1, Supporting Information). Therefore, the formation of terraces in the 3.5 nm thick film, exhibiting a wider dispersion in \( S \) values, can be attributed to the overlapping of the strain field in regions with locally enhanced MD densities.

### 2.3. Strain Effects on Surface Currents

Before considering the local effect of buried MDs on surface currents, we will take into consideration the bulk transport...
behavior of the films. Figure 5a depicts standard four-point electrical resistivity measurements as a function of the thickness of the films. As seen in the figure, room temperature resistivity values decrease as the film thickness, thereof the relaxation level as manifested by the variation in the \(c/a\) lattice parameter ratio, increases. The temperature dependence of the resistivity of the different films, on the other hand, indicates bulk-like insulating behavior for the 2 nm and 3.5 nm thick films, and metallic behavior for larger thicknesses (6 and 14 nm), see Figure S2 (Supporting Information). As far as the surface electrical properties of the films are concerned, however, current maps indicate local resistivity fluctuations that appear associated with the formation of MDs. Starting with the 3.5 nm thick film, Figure 5b shows an AFM image corresponding to the \(I(x,z)\) current map shown in Figure 5c. Despite its bulk insulating behavior, this film exhibits surface metallic behavior. Current enhancements are clearly seen as dark contrasts decorating surface steps, which are attributed to the extended tip-surface contact area along their ledges. Within the terraces, brighter lines of depressed current are also observed along the in-plane \(<100>\) directions. Both, topographic and conduction images are correlated with the underlying MD network. For the 6 nm thick films, the topographic ridge/terrace pattern is no longer present (Figure 5d and Figure S1, Supporting Information) and exhibits unit cell height steps (Figure 5f), the corresponding current map still exhibits current depressions along lines parallel to the in-plane \(<100>\) directions, as clearly seen in Figure 5e.

The influence of strain on the magnetotransport properties ABO\(_3\) perovskite compounds is intimately correlated to tilt and distortion processes of the MnO\(_6\) octahedral framework. Strain affects magnetotransport properties by acting on different mechanisms at a microscopic level. First strain affects both Mn–O–Mn bond angle and the Mn–O bond length, thus modifying the strength of the double exchange ferromagnetic (DEF) interactions. For the same reasons strain also affects antiferromagnetic (AF) superexchange interactions. In addition, strain may introduce an orbital bias since in-plane compressive or tensile strains may promote selective \(d\ 3z^2-r^2\) or \(d\ 3x^2-y^2\) orbital occupancy, respectively. Therefore, as a first approximation, an elongation of the Mn–O distances or a decrease of the Mn–O–Mn bond angle would promote a reduction of the strength of DEF interactions and therefore, a reduction of the ferromagnetic Curie temperature, \(T_C\), and an increase of electrical resistivity. On the contrary, the reduction of Mn–O distances or the straightening of the Mn–O–Mn bond angle promotes the strengthening of DEF interactions and a reduction of resistivity. The observation of enhanced conductivity at (100)-type twin walls in LSMO/STO thin films, which are submitted to a severe compressive strain, indeed

Figure 5. a) Room temperature thickness dependence of the resistivity of the LSMO/LAO films. b,d) Topography and c,e) simultaneous current maps for 3.5 and 6 nm thick films, respectively. f) Line profile along the segment indicated in panel (d), the step height corresponds to one unit cell (\(\approx 4\ \text{Å}\)). Dashed lines, black (c)/white (e), in the current maps serve as a guide to the eye to highlight the dislocation network on the film surface.
supports these arguments.\[49\] Nevertheless, this scenario may be strongly affected by selective orbital occupancy and AF superexchange interactions. The balance between these competing effects is controlled by the ratio between perpendicular and in-plane lattice parameters $c/a$, reflecting the degree of tetragonal distortion of the structure.\[50\] In such scenario, increasing $c/a > 1.0$ works against the metallic ferromagnetic behavior and would promote an increment of the resistivity. Since according to our analysis above, the behavior of the present films is well described by the continuum isotropic elasticity theory, the influence of rigid octahedral tilting mechanisms can be safely neglected, which otherwise would manifest as noticeable anomalies in the behavior of lattice parameters.\[7\] Accordingly, the image contrast in the AFM current maps may be analyzed in terms of local $c/a$ values, directly governed by the residual misfit strain, $(\varepsilon_{xx}+\varepsilon_{zz})(x)$ shown in Figure 4a. The lateral modulation of the $c/a$ ratio at the film surface is given by $c/a(x) = (\varepsilon_{xx}(x)+1)/[(\varepsilon_{xx}+\varepsilon_{zz})(x)+1]$, where $\varepsilon_{xx}$ is given by Equation (2), and $\varepsilon_{zz}$ is solely determined by the outward relaxation determined by the Poisson's effect, $\varepsilon_{zz} = 2\nu\varepsilon_{xx}/(1-\nu)$. In the case of the 6 nm thick film, using the Poisson's ratio derived for the 2 nm thick film, $\nu = 0.35$ (see above), a $c/a$ ratio of $\approx 1.03$ can be estimated at the locus of the dislocation ($\varepsilon_{xx} = -0.013$) while $c/a \approx 1.0$ in a large area between dislocations ($\varepsilon_{xx} = 0$). As a result, the metallic character is expected to be depressed at the dislocations that would exhibit a higher resistivity. Thus, the pattern observed in current maps is a close mimic of the dislocations pattern. The very same reasoning may be applied to explain the low conducting paths observed on the surface of the 3.5 nm thick film. In this case we find $c/a = 1.05$ ($\varepsilon_{xx} = -0.020$) above the dislocation lines. Experimental determinations of the $c/a$ ratio from the GPA derived strain maps indeed support the above behavior. Figure 6a shows the $c/a(x)$ dependence at two different levels above the dislocation analyzed in Figure 1c,d, corresponding to the 3.5 nm thick film. The blue and red curves are taken close to the film surface and at half way from the buried dislocation core, as indicated in Figure 6b. It can be clearly observed that the $c/a$ ratio locally increases above the dislocation, reaching values closely similar to those derived from linear elasticity. It can be also observed that the average $c/a$ level increases toward the film surface, which can be attributed to an elastic relaxation perpendicular to the film surface.

Identifying the strained regions as less conducting is also in agreement with the insulating behavior observed in the 2 nm thick film, prior to the appearance of dislocations. In those films, $c/a = 1.06$ ($\varepsilon_{xx} = -0.023$), i.e., they are expected to lie well within an antiferromagnetic insulating phase.\[6\] Above the onset of plastic relaxation, this analysis yields a view of partly relaxed LSMO/LAO thin films as a conducting compressed matrix, decorated with nanometric paths of higher resistance material aligned with the in-plane $<100>$ directions coinciding with the positions of the buried MDs, in agreement with experimental $I(x,y)$ maps. As shown above, the topological distribution of strained, high-resistivity regions, throughout the volume of the films varies with the film thickness. Just after the onset of plastic relaxation, the MD density is low, but their associated strains occupy a significant fraction of the film volume owing to its reduced thickness. Thus, in the 3.5 nm thick film, the average bulk insulating behavior can be understood by considering that the connectivity between metallic regions is below the percolating threshold. At larger thicknesses, the increased dislocation density promotes a higher average level of relaxation consistent with the metallic behavior observed in the 6 nm thick film.

3. Conclusion

To conclude, the strain field of MDs in a complex oxide heterostructure introduces a lateral modulation of the chemical potential and the bandwidth-independent properties at the free surface of the film. In particular, the present experiments shed light on the structural mechanisms underpinning the topographic and electrical conduction patterning of the surface of LSMO/LAO thin films. It is shown that, even if perovskite thin films may relax misfit strains by combining octahedral distortions and octahedral tilting which may eventually elude the formation of...
4. Experimental Section

Films: High-quality films with (100) orientation and thicknesses of 2, 3.5, 6, and 14 nm were epitaxially grown on a biaxial compressive strain of $t_0 = -2.3\%$ on LAO substrates by magnetron sputtering as reported elsewhere.\[16\] Conducting Atomic Force Microscopy: Local atomic conductivities were measured using a commercial conductive CrPt-coated Si tips mounted on cantilevers with $k = 40$ Nm$^{-1}$ (BudgetSensors). The lateral resolution of the technique is, in principle, limited by the tip radius to $\approx 10-20$ nm. In the employed setup, the sample was grounded and the voltage was applied to the tip. An external $I-V$ converter (Stanford Research Systems) was used to provide access to a wide range of compliance currents (1 pA to 1 mA).

High-Resolution X-Ray Diffraction: The lattice parameters of the films were determined from HR-XRD using a four-angle goniometer and primary optics consisting of a parabolic mirror and a $4 \times$ Ge(220) asymmetric monochromator (X’Pert Pro MRD-Panalytical). The in-plane lattice parameters were determined in the same equipment with a parabolic mirror in the incidence beam optics, fixed grazing angle of $0.5^\circ$ on the sample, and parallel plate collimator in the diffracted optics.

Transmission Electron Microscopy: Thin foil specimens were prepared by conventional cutting–gluing–grinding procedures, followed by Ar milling at a grazing incidence down to perforation. Cross-section atomic resolution images (HRTEM) were obtained at 200 kV using the field emission gun FEI Tecnai F20 S/TEM and the Cs-corrected Tecnai F20 electron microscopes. The projected strain distribution on the plane of the film was directly imaged by LAADF microscopy,\[32\] using a field emission gun Tecnai F20 S/TEM electron microscope.

Orientation Contrast Scanning Electron Microscopy: Orientation contrast SEM images\[53\] were obtained in a QUANTA FEI 200 FEG-ESEM electron microscope in order to determine dislocation densities over large areas.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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