



Moiré engineering of electronic phenomena in correlated oxides

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Moiré engineering has recently emerged as an effective approach to control quantum phenomena in condensed matter systems^{1–6}. In van der Waals heterostructures, moiré patterns can be formed by lattice misorientation between adjacent atomic layers, creating long-range electronic order. Moiré engineering has so far been executed solely in stacked van der Waals multilayers. Here we describe electronic moiré patterns in films of a prototypical magnetoresistive oxide, $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$, epitaxially grown on LaAlO_3 substrates. Using scanning probe nanoimaging, we observe microscopic moiré profiles attributed to the coexistence and interaction of two distinct incommensurate patterns of strain modulation within these films. The net effect is that both the electronic conductivity and ferromagnetism of $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ are modulated by periodic moiré textures extending over mesoscopic scales. Our work provides a potential route to achieving spatially patterned electronic textures on demand in strained epitaxial materials.

Moiré patterns emerge from overlaying two sets of mildly dissimilar periodic motifs. In two-dimensional van der Waals systems, moiré patterns have been achieved by stacking two-dimensional atomic layers with slightly incommensurate periodicities and/or small twist angles⁷. Fascinating properties have emerged in two-dimensional moiré systems, including superconductivity and Mott insulating states in magic-angle graphene superlattices^{1,2}, moiré excitons in transition metal dichalcogenide heterobilayers^{3–5} and topological conducting channels in twisted bilayer graphene⁶. However, moiré-type electronic modulations have never been experimentally demonstrated or theoretically proposed in correlated transition metal oxides (CTMOs), which are known to host rich interactions between charge, spin, lattice and orbital degrees of freedom^{8,9}. Herein, we demonstrate that moiré engineering is applicable beyond van der Waals materials and report mesoscale spatial moiré modulations of coupled conductivity and ferromagnetism in strained thin films of a correlated oxide.

A universal property of CTMOs is the competition and coexistence of multiple order parameters that spontaneously give rise to spatially textured physical properties¹⁰. These emergent electronic textures are particularly prominent in manganites, including $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ and related materials^{11–14}. However, deterministic

creation of spatially ordered patterns remains a daunting task. Here, we utilize interactions between two distinct sources of periodic strain modulation in epitaxial $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ (LSMO) films grown on LaAlO_3 (LAO) substrates. When these two modulations are collocated in real space, they coproduce moiré-like energy landscapes that locally modulate conductivity and ferromagnetism.

We investigated 20-nm-thick LSMO films on LAO substrates (see Methods for details). The high crystalline and epitaxial quality is confirmed by a combination of X-ray diffraction (XRD) and cross-sectional transmission electron microscopy (TEM) (Extended Data Fig. 1). Temperature-dependent resistivity and magnetic moment reveal a characteristic second-order phase transition from ferromagnetic metal to paramagnetic metal at $T_C \sim 340$ K (Fig. 1a). This T_C is reduced compared with the bulk value (~ 370 K; ref. ¹⁵), attesting to the large in-plane compressive strain imposed by the substrate¹⁶. We harnessed epitaxial control to define the local compressive strain patterns ultimately giving rise to moiré textures. First, we note that LSMO films on LAO possess highly oriented rhombohedral twin domains^{17–21} that stem from local relaxations of substrate-imposed shear strain (see Fig. 1b, Extended Data Fig. 3 and Supplementary Note 2). As schematically shown in Fig. 1c, such a periodic structure results in a striped strain modulation^{18,21}, which we designate as domain stripes (DSs). On the other hand, surface miscut steps in the LAO substrate (see Fig. 1d,e and Extended Data Fig. 4) serve as another source of periodically modulated strain in LSMO. The resultant periodic strain modulation in LSMO, which we name miscut stripes (MSs), does not necessarily align with any specific crystal axis but depends strictly on the local miscut surface of the substrate²² (see Supplementary Note 3). The spatial coexistence of DS and MS modulations yields moiré landscapes with domain sizes governed by the periodicities and relative angles of the two constituent stripe motifs (Fig. 1f).

Nano-optical imaging provides direct evidence for the promise and possibility of moiré strain engineering. We utilized infrared scattering-type scanning near-field optical microscopy (s-SNOM) to visualize nanoscale moiré phenomena in strained LSMO/LAO (see Methods for details). The amplitude of the nano-infrared signal demodulated at the second harmonic of the tip tapping frequency (S_2) provides a local probe of the optical conductivity with ~ 25 -nm

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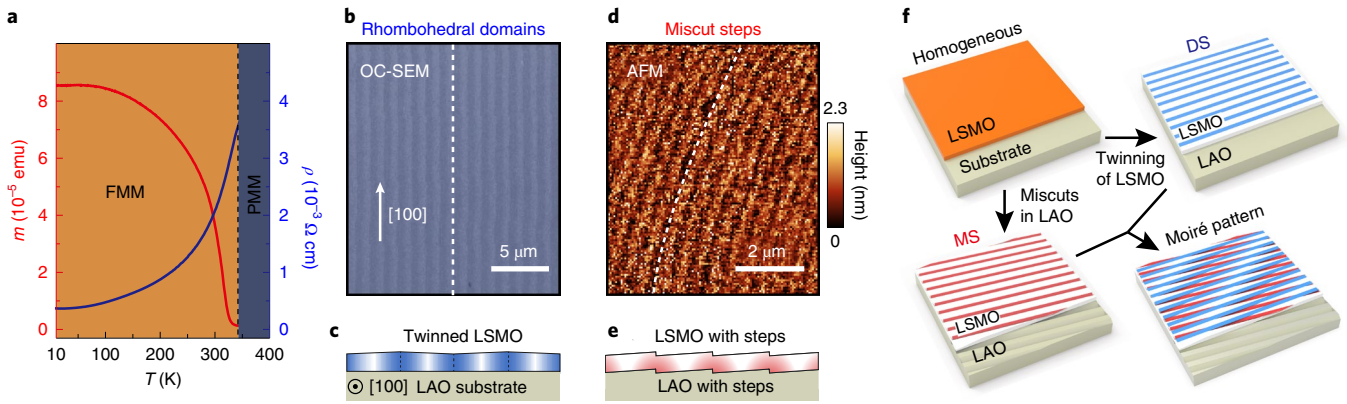


Fig. 1 | Two types of periodic strain modulation in LSMO thin films. **a**, Temperature-dependent resistivity ρ and magnetic moment m of an LSMO thin film under study. The magnetic moment was measured at 100 Oe on warming after cooling the sample in a 100-Oe field. The black dashed line indicates T_C . FMM, ferromagnetic metal; PMM, paramagnetic metal. **b**, Orientation-contrast scanning electron micrograph (OC-SEM) image of an LSMO thin film, showing surface rhombohedral domains with a width of ~ 590 nm aligned along the $[100]$ direction. **c**, Schematic of the rhombohedral-domain-induced strain modulation in an LSMO-LAO cross-section. **d**, AFM image of an LSMO thin film, showing topographic surface changes due to crystal miscut steps in the LAO substrate. The period is ~ 500 nm. White dashed lines in **b** and **d** indicate the directions of the rhombohedral domains and miscut steps, respectively. **e**, Schematic of the miscut-step-induced strain modulation in an LSMO-LAO cross-section. **f**, Schematic demonstration of how the spatial coexistence of DSs and MSs yields the combined moiré patterns comprising regions of local commensuration and incommensuration.

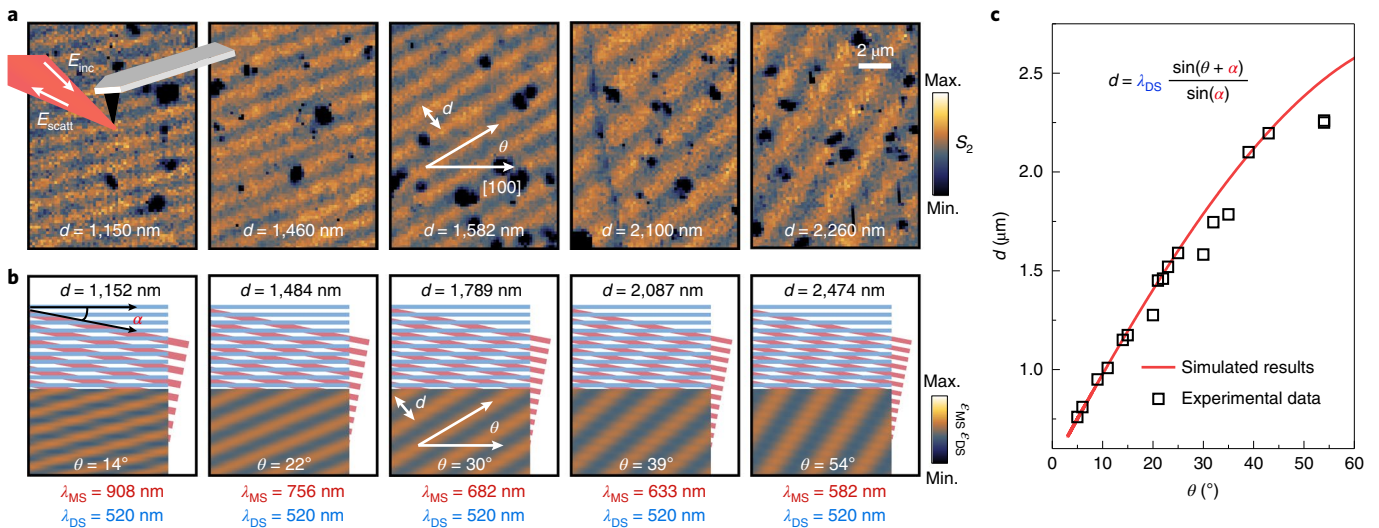


Fig. 2 | Electronic moiré patterns in LSMO films. **a**, Nano-infrared maps of S_2 obtained at the infrared wavelength of $10.7 \mu\text{m}$ and collected in different regions of a representative LSMO/LAO film. **b**, Top: simulations of the moiré patterns formed by superimposing two periodic striped motifs. Blue, DSs; red, MSs. From left to right, the λ_{MS} are 908 nm, 756 nm, 682 nm, 633 nm and 582 nm, respectively, while λ_{DS} and the twist angle between DSs and MSs are fixed at 520 nm and 11° , respectively. Bottom: simulations generated by multiplying the two striped patterns using the same parameters as in the top half. **c**, The moiré pattern periodicity as a function of θ .

spatial resolution^{23,24}. In Fig. 2a, we display maps of the two-dimensional nano-infrared contrast, revealing periodic modulations of the local electronic response in LSMO/LAO. Notably, the periodicity d is revealed to be considerably larger than that of either the DSs or MSs. Moreover, d appears to be governed by the orientation of these patterns with respect to the $[100]$ direction of the LAO substrate (also see Extended Data Fig. 5). These systematic trends point to a unified underlying mechanism for the formation of electronic patterns that we link to the emergent moiré pattern of combined periodic strains.

We now proceed to elucidate the rich real-space structures of conductivity observed in our strained films. We first note that the two sets of overlaid unidirectional stripes with relatively small spacings readily produce longer-period textures (top panels in

Fig. 2b). Microscopic strain fields ϵ_{DS} and ϵ_{MS} that vary sinusoidally in space present a realistic scenario for our films. Here we associate ϵ_{DS} with the DSs and ϵ_{MS} with the MSs introduced in Fig. 1. As demonstrated in the bottom panels of Fig. 2b, we were able to reproduce the key trends in the images in Fig. 2a by assuming that the observed textures are a direct manifestation of the product $\epsilon_{DS} \cdot \epsilon_{MS}$ of these two sinusoidal strain fields. This product term naturally stems from the nonlinear relationship between the strain and the conductivity, which originates in the strain-induced nonlinear T_C modulation in LSMO^{16,25–27} (detailed in Supplementary Note 4). Our experimental result attests to the large magnitude of nonlinear coefficient for the cross-term $\epsilon_{DS} \cdot \epsilon_{MS}$, which originates from the cubic (O_h) point symmetry of Mn in the cubic perovskite structure

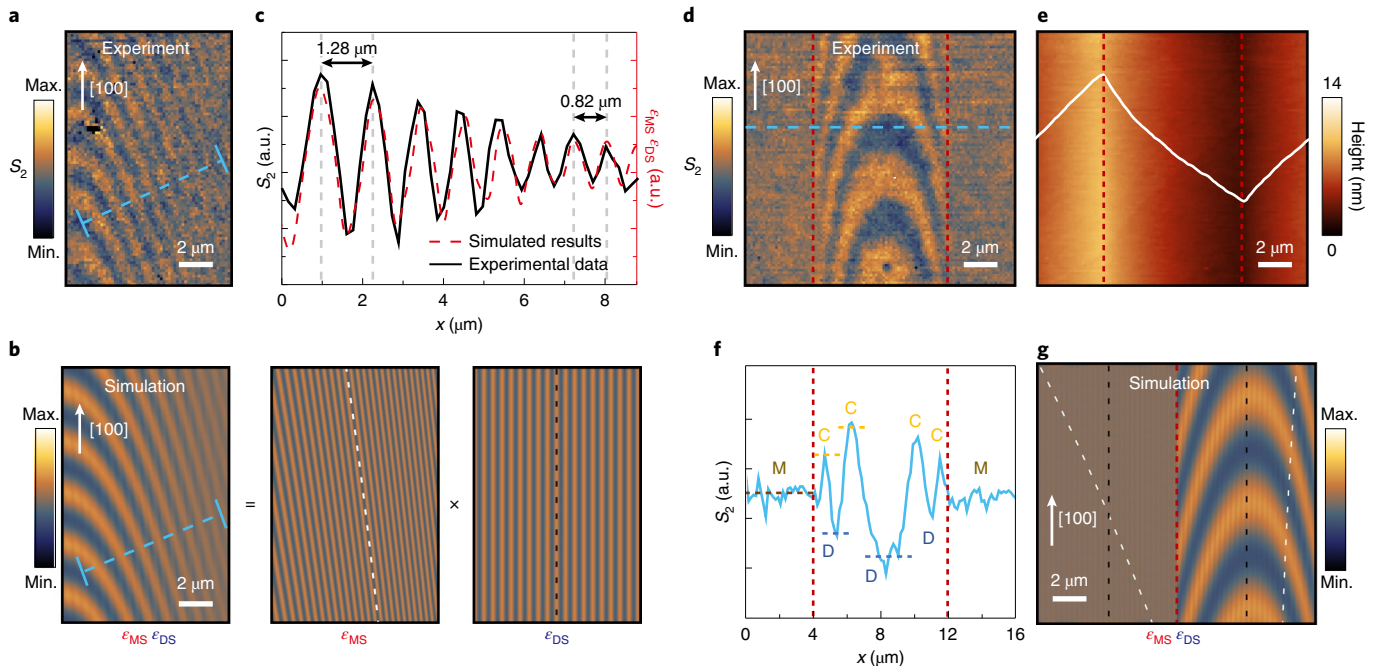


Fig. 3 | Curved electronic moiré patterns in LSMO films. **a, b**, Infrared near-field image of a curved moiré pattern (**a**) and the corresponding simulation (**b**). The simulation is generated by multiplying two periodic striped patterns representing MSs and DSs. The white and black dashed lines in **b** indicate the MS and DS orientations, respectively. **c**, Line profiles from the blue dashed lines in **a** and **b**, exhibiting high consistency between the experimental and the simulated contrast. a.u., arbitrary units. **d, e**, Infrared near-field image and corresponding AFM image, respectively, showing alternating moiré and non-moiré regions across the LAO twin boundaries (indicated by red dashed lines). The white solid line in **e** is the AFM height profile. **f**, Line profile of the nano-infrared contrast along the blue dashed line in **d**. The different signal levels are marked 'C', 'D' and 'M', which represent constructively strained, destructively strained and mixed strained regions, respectively. **g**, Simulation of the image in **d** with the moiré pattern only visible on the right-hand side. Note that the MSs (orientation indicated by white dashed lines) change orientation across the LAO twin boundary (red dashed line), while the DSs (orientation indicated by black dashed lines) are consistently along the LAO [100] direction. The simulation details are shown in Extended Data Fig. 8c.

as well as the large Jahn–Teller effect due to strong electron–lattice coupling in LSMO²⁵. We note that, in our simulated image, a local Gaussian average filter with radius ~ 100 nm is applied to account for the finite spatial resolution of the near-field probe and possible local strain relaxation (Extended Data Fig. 6).

The concurrent evolution of the moiré fringe d and angle θ (defined as the angle between the orientation of fringes and the LAO [100] direction) can be theoretically obtained by multiplying two sinusoidal waves with wavevectors $\mathbf{k}_{\text{DS}} = (0, \frac{2\pi}{\lambda_{\text{DS}}})$ and $\mathbf{k}_{\text{MS}} = (\frac{2\pi \sin(\alpha)}{\lambda_{\text{MS}}}, \frac{2\pi \cos(\alpha)}{\lambda_{\text{MS}}})$, where λ_{DS} and λ_{MS} are the DS and MS periodicities, respectively, and α is the twist angle between the DSs and MSs. The resultant moiré pattern yields $\theta = \arccot(\frac{\lambda_{\text{MS}} - \lambda_{\text{DS}} \cos(\alpha)}{\lambda_{\text{DS}} \sin(\alpha)})$ and $d = \frac{2\pi}{|\mathbf{k}_{\text{DS}} - \mathbf{k}_{\text{MS}}|} = \frac{\lambda_{\text{DS}} \sin(\theta + \alpha)}{\sin(\alpha)}$. We found that the d – θ relation shown in Fig. 2a and Extended Data Fig. 5 can be reproduced by solely adjusting the λ_{MS} , whose gradual change is expected across the 1×1 mm² film due to a long-range variation in the surface miscut angle of the substrate and consequently in the MS periodicity. As shown in Fig. 2b,c, we found excellent agreement between experimental images and theory by monotonically varying λ_{MS} from 908 nm to 582 nm while keeping λ_{DS} and α constant. This agreement verifies that the spatial modulations in optical conductivity observed in the LSMO/LAO samples do indeed arise from the synergistic effect of DSs and MSs. We note that similar moiré fringes were not found in the surface topography of LSMO corecorded with the near-field imaging using atomic force microscopy (AFM), which suggests that the strong nonlinear coupling of optical conductivity to the local strain is an essential ingredient for deterministic occurrence of observed moiré patterns.

Apart from the unidirectional moiré fringes in Fig. 2, more complex moiré patterns can be obtained (Extended Data Fig. 7). For example, we observed curved moiré fringes in another LSMO/LAO sample of the same ~ 20 -nm thickness (Fig. 3a). In Fig. 3b we show that such curved moiré patterns can also be perfectly reproduced by the product $\epsilon_{\text{DS}} \cdot \epsilon_{\text{MS}}$ under the assumption of MSs with a slight curvature smaller than $\sim \frac{1}{100 \mu\text{m}}$ (Supplementary Note 5). The line profiles thus obtained reveal an astonishing consistency between experiments and simulations, where the amplitude contrast of variations in optical conductivity resolved by our nanoimaging decays with decreasing periodicity (Fig. 3c; see also Supplementary Information Fig. 1). This consistency further confirms the validity of our interpretation. Furthermore, the variations of the orientation and curvature of the MSs offer a unique opportunity to realize a diverse range of electronic moiré textures with different spatial configurations (Extended Data Fig. 8).

In Fig. 3d, another example of a curved moiré pattern is presented. The near-field line profile (Fig. 3f) demonstrates clear contrast between peaks and valleys of optical conductivity in the moiré pattern regions. On the other hand, regions without a clear moiré pattern reveal optical conductivity approximately equal to the average of that of the moiré fringes. Note that the borders between moiré and non-moiré regions coincide with the twin boundaries of the LAO substrate (red dashed lines in Fig. 3d–f). In Fig. 3e, the topographic variation up to 10 nm in height originates from the LAO substrate twin structures^{28,29} (Extended Data Fig. 2a and Supplementary Note 1). No visible moiré-like topographic modulation can be seen. Here we give a phenomenological interpretation: the miscut steps in LAO possess markedly different periodicities, orientations and even curvatures on either side of the underlying

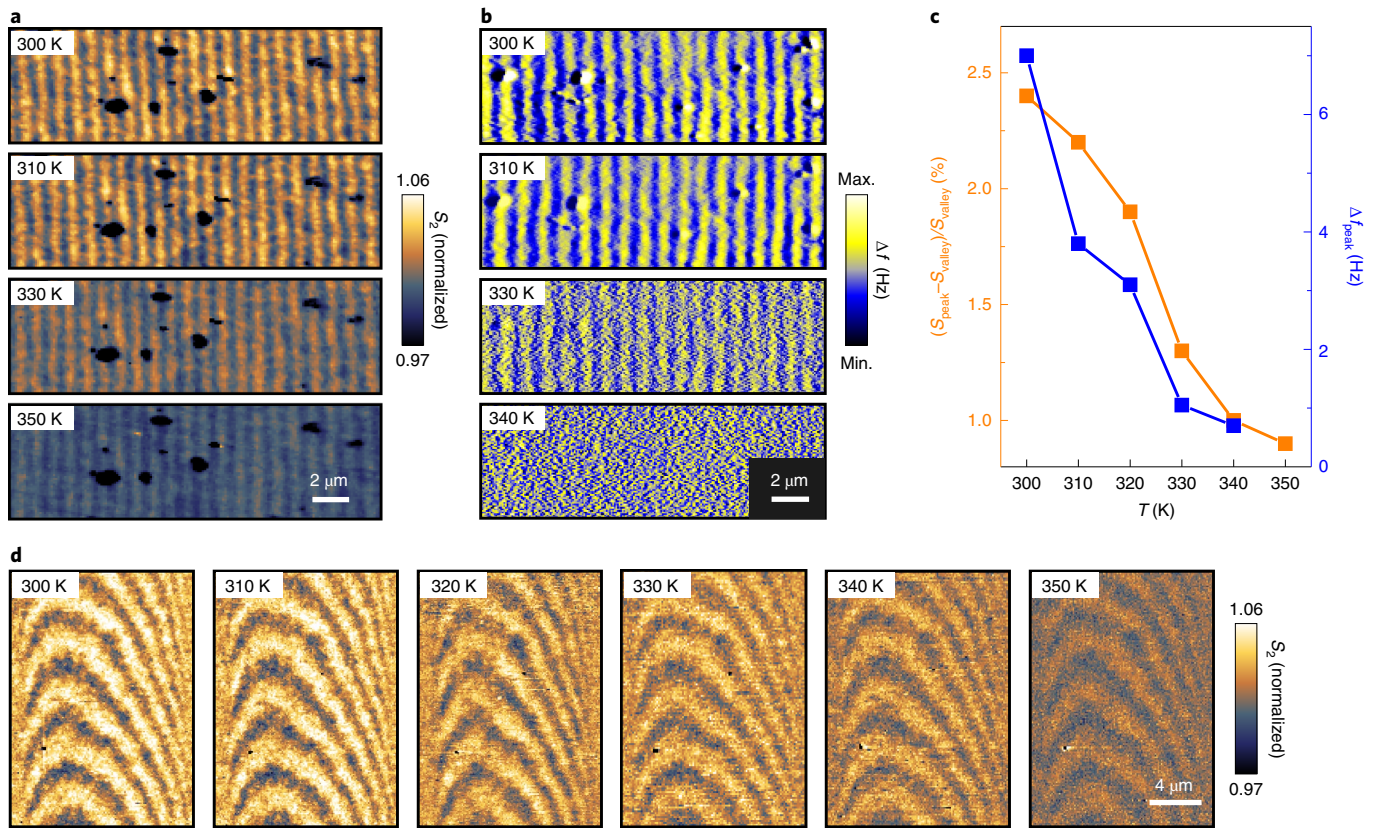


Fig. 4 | Temperature-dependent evolution of electronic and magnetic moiré patterns. **a, b**, Near-field and MFM images, respectively, of a moiré pattern measured at elevated temperatures. **c**, Temperature-dependent near-field contrast $(S_{\text{peak}} - S_{\text{valley}})/S_{\text{valley}}$ and MFM peak-frequency shift Δf_{peak} extracted from images in **a** and **b**, respectively. **d**, Near-field images of the curved moiré pattern obtained at different temperatures. All of the near-field images are normalized to dark fringes to demonstrate the relative contrast fading with increasing temperature.

LAO twin boundaries (Extended Data Fig. 9). As simulated in Fig. 3g (detailed in Extended Data Fig. 8c), to the right of the LAO twin boundary, the small twist angle between DS and MS leads to a relatively large moiré pattern periodicity that can be observed under near-field imaging. To the left of the LAO twin boundary, the large twist angle is expected to produce a moiré pattern with a much smaller periodicity, of the same order as the DSs or MSs. As demonstrated in Fig. 3c and Supplementary Information Fig. 1, smaller periodicity yields a smaller infrared near-field intensity contrast. The spatial features are therefore indistinguishable under the near-field mapping due to minimal conductivity variation between the electronic domains. This is supported experimentally by the average signal level detected for the non-moiré regions in Fig. 3f.

Temperature-dependent s-SNOM and magnetic force microscopy (MFM) measurements provide further insight into the interdependence between electronic and magnetic moiré textures in strained LSMO. As a hallmark feature of the colossal magnetoresistive manganites, the microscopic mechanism of electrical conductivity in LSMO is intimately tied to the onset of long-range ferromagnetic order⁹, and here we report moiré-like modulations of that dependence borne out and resolved at the mesoscale. Clear fringes in moiré conductivity are observed with a periodicity of $\sim 1.05 \mu\text{m}$ at room temperature, as shown in the near-field images of Fig. 4a. Figure 4b shows the MFM images acquired in roughly the same region, where clear frequency-shift contrast with the periodicity of $\sim 1.13 \mu\text{m}$ is observed at 300 K, demonstrating the moiré modulations of local ferromagnetism. Increasing the temperature prompts a decrease in both the nano-infrared contrast and magnetic response (Fig. 4c). This latter observation implies a positive

correlation between the conductivity and the local ferromagnetic moment, consistent with the established phase diagram of LSMO¹⁵. The observed magnetic moiré fringes disappear at the ferromagnetic–paramagnetic transition temperature of 340 K (Fig. 4b and Supplementary Information Fig. 3), while the electronic counterpart persists with a greatly reduced nano-infrared contrast up to all measurable temperatures (Fig. 4a). This may suggest that two distinguishable sets of ferromagnetic metal states with different strain environments transfer to two paramagnetic metal states at T_C . Above T_C no magnetization persists, but detectable near-field contrast is still present due to the local strain modulation. The temperature evolution of a curved moiré pattern is also shown in Fig. 4d. It is evident that the near-field optical contrast of the moiré pattern decays substantially at higher temperatures. Such an observation further validates that the electronic and magnetic properties of the moiré patterns are closely related to the T_C of LSMO/LAO thin films.

Finally, we wish to point out that the individual DS and MS stripes are barely observable under s-SNOM, but their combined electronic effects nevertheless manifest clearly through the moiré pattern formation (Extended Data Fig. 10). This ‘magnified’ electronic response may be potentially useful for identifying ‘hidden’ textures of strain, electronic or magnetic distribution in CTMO thin films, where individual traits are difficult to identify separately.

In summary, our work reports the ordered electronic and magnetic mesoscale patterns in LSMO, demonstrating the feasibility of producing and visualizing surface moiré patterns that yield optical contrast. In the large family of CTMO, there are many oxide systems that host substantial nonlinear strain coupling, for example those

with a cubic or pseudocubic crystal structure, and a relatively large Jahn–Teller effect²⁴. Our novel approach thus provides a strategy for manipulating complex electronic orders in strongly correlated quantum materials such as NdNiO₃^{22,30} or SrTiO₃³¹, where long-wavelength electronic and lattice modulations similar to those exploited here have been reported. Beyond the long-wavelength moiré patterns, it is also interesting to explore similar effects at shorter length scales. Materials with competing phases or proximate to a quantum phase transition can be tested with high priorities (for example the interplay between the stripe and superconducting phases in high-*T_c* cuprates is affected by strain).

Online content

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Methods

Sample fabrication and characterization. LSMO thin films of varying thicknesses were grown on the LAO(001) substrates at 700 °C via pulsed laser deposition. A KrF excimer laser was used with an energy density of 5.5 J cm^{-2} and a 3-Hz repetition rate. The O_2 pressure was maintained at 40 Pa during growth. The as-grown films were then in situ annealed for 0.5 h under the deposition conditions, followed by ex situ annealing in a tube furnace at 780 °C in flowing O_2 . The lattice parameters and crystalline quality of the films were characterized by XRD (Panalytical X'Pert PRO MRD) and cross-sectional TEM at room temperature, with typical results for the 20-nm sample shown in Extended Data Fig. 1. Our films are found to be of good crystalline and epitaxial quality. The relatively large full-width at half-maximum of the rocking curve of the LSMO 002 reflection indicates the appearance of bending or twinning of the 002 crystal plane, which is consistent with the lattice strain effect proposed above for the moiré pattern. The magnetic properties were analysed using a Quantum Design SQUID-VSM system. The electrical transport properties were measured using the four-probe method in a Quantum Design PPMS system.

Infrared s-SNOM. Measurements were conducted with two different s-SNOM apparatuses, yielding nearly identical results. Monochromatic light of wavelength $\sim 11 \mu\text{m}$ ($\sim 10.7 \mu\text{m}$ for the other s-SNOM) was focused onto an AFM probe with tip apex radius $\sim 25 \text{ nm}$. The AFM probe was operated in tapping mode with oscillation frequency $\Omega \sim 250 \text{ kHz}$ and amplitude $A \sim 50 \text{ nm}$ ($\sim 80 \text{ nm}$ for the other s-SNOM). Backscattered light from the tip-sample ensemble interfered with the reference light to yield phase-resolved infrared near-field detection. Lock-in demodulation at higher harmonics of Ω could largely eliminate undesired background and preserve the genuine local near-field interaction emanating from the confined area below the tip apex. In our measurements, the amplitudes of the nano-infrared signal demodulated at the second/third/fourth harmonic of the tip tapping frequency ($S_2/S_3/S_4$) showed essentially identical results, and the S_2 signal was finally presented for better signal-noise ratio.

MFM. MFM measurements were performed under zero magnetic field with a home-made MFM. Using electron-beam deposition, the tip was coated with a trilayer film of 5 nm Ti, 50 nm Co and 5 nm Au, which was subsequently magnetized with a permanent magnet along the tip axis. The MFM images were obtained in the frequency mode, in which the frequency shift Δf ($\Delta f = f - f_0$) was 'tracked' by a commercial phase-locked loop. A negative frequency shift indicates an attractive tip-sample interaction ($\Delta f < 0$), while a positive frequency shift indicates a repulsive tip-sample interaction ($\Delta f > 0$).

OC-SEM. The rhombohedral domains in the LSMO/LAO samples were characterized using OC-SEM imaging, with misorientation between different domains represented by different contrast levels. The measurements were conducted in a field emission scanning electron microscope (ZEISS GeminiSEM 500) using the in-lens mode. The typical values of the acceleration voltage, objective aperture size and working distance were 1 kV, $120 \mu\text{m}$ and 5 mm, respectively.

Data availability

The data represented in Figs. 1–4 are available as Source Data 1–4. All other data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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Author contributions

M.L., C.Z. and D.N.B. designed and supervised the work. X.C. and X.F. performed the s-SNOM measurements with assistance from Z.N., S.X., D.W. and H.Z.; L.L., N.Z. and H.X. fabricated the samples and performed the XRD, magnetic and transport characterizations; T.G. and Q.L. performed the MFM measurements; X.C., L.L., X.F., D.N.B., M.L. and C.Z. analysed the data and wrote the manuscript. A.S.M., A.J.M. and Z.L. contributed to data interpretation and presentation. All authors contributed to the scientific discussion and manuscript revisions.

Competing interests

The authors declare no competing interests.

Additional information

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