Nano-textured phase coexistence in the correlated insulator V_2O_3

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Abstract:

While the insulator-metal transition remains among the most studied phenomena in correlated electron physics, its interplay with spatially textured phase coexistence at the meso- and nano-scales remains poorly explored. Here we reveal real-space evolution of the low temperature insulator-metal transition in a V_2O_3 thin film imaged at high spatial resolution by cryogenic near-field infrared microscopy. In the vicinity of insulator-to-metal temperature (~ 160 - 180 K) we resolve spontaneously nano-textured coexistence of metal and correlated (Mott) insulator phases associated with percolation and an underlying structural phase transition. Augmented with macroscopic temperature-dependent x-ray diffraction measurements of the same film, a quantitative analysis of nano-infrared images acquired across the transition suggests decoupling of electronic and structural transformations. Persistent low-temperature metallicity is accompanied by

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unconventional critical behavior through the film's first-order insulator-metal transition, suggestive of Coulomb frustration.

Manuscript Text:

The subtle interplay of charge, spin, and orbital degrees of freedom in complex correlated oxides is known to result in strongly inhomogeneous phases¹. In several families of correlated oxides including cuprates^{2,3} and manganites^{4,5,6}, experimentally observed emergent real-space patterns provide fundamental clues to the principles underlying exotic behavior of these systems. The real-space self-organization of electronic phases remains largely unexplored in many classes of materials exhibiting a first-order insulator-metal transition (IMT), attributable mostly to the absence of real-space probes capable of imaging the highly insulating side of the transition with nano-scale resolution. Enabled by nano-resolved infrared (nano-IR) imaging⁷, we demonstrate in this article how a self-organized real-space electronic nanotexture can underly more than five orders of magnitude in increased resistivity through the IMT of V_2O_3 . Furthermore, we resolve hitherto unreported characteristics of this correlated electron system, demonstrating that stabilized phase coexistent can provide a unique window onto the interactions governing an insulator-metal transition that was previously considered monolithically first-order.

The IMT in V_2O_3 from paramagnetic metal (PM) to low temperature ($T \sim 150$ K) antiferromagnetic insulator (AFI) arises from thermally perturbing the delicate competition between kinetic energy and mutual Coulomb repulsion among conduction electrons^{8,9}, leading to abrupt electron localization and a 0.7 eV band-

gap¹⁰. These features appear intimately tied to a structural phase transition (SPT) from a hexagonal (corundum) lattice to a lower symmetry monoclinic structure⁸. In recent years, increasingly refined studies have revealed unforeseen aspects of this transition^{11,12,13}, hypothesizing a more subtle phase diagram where real-space phase inhomogeneities play an essential role. The IMT and SPT have been found to decouple in the related compound VO₂, whereby a strongly correlated monoclinic metal emerges between low temperature monoclinic insulating and high temperature rutile metallic phases^{14,15,16}. Real-space phase separation amidst this IMT results in a resistive transition governed by percolation. However, the exact role of Mott physics in VO₂ remains hotly debated, calling for a critical examination of real-space features amidst the IMT of V₂O₃ at low-*T*, where a localization-delocalization mechanism is more conclusively at work^{9,17 18,19}.

We developed cryogenic infrared near-field (nano-IR) imaging to assess nano-scale phase inhomogeneity emerging through the classic IMT in V₂O₃. To explore phase coexistence amidst the transition, we image a high-quality V₂O₃ thin film, revealing a real-space nanotexture reflective of interactions governing the IMT. At temperatures of phase coexistence, our local nano-optical probe first resolves bi-directional stripes of the correlated insulator phase percolating through the paramagnetic metal, whereas at temperatures below the transition metallic droplets persist in an otherwise insulating background. Nevertheless, macroscopic x-ray diffraction (XRD) of the same film indicates a virtually complete structural phase transition from corundum to monoclinic structure at these temperatures. Our nano-IR imaging therefore reveals V₂O₃ can support a monoclinic metal phase.

Characterization of evolving real-space correlations and critical scaling among electronic inhomogeneities suggest that Coulomb frustration may be operative in the stability of monoclinic metallicity.

Scattering-type scanning near-field optical microscopy (s-SNOM) enables sub-diffractive imaging of surface optical properties^{20,7} at variable temperatures^{21,22}, with a resolution strictly limited only by the geometric probe sharpness. We present 25 nm-resolved images of the locally back-scattered near-field amplitude (abbreviated to *nano-IR signal*, or *S*) collected at low temperatures (Fig. 1a) by a newly developed cryogenic near-field scanning optical microscope (further details in Supplementary Information [SI] and fig. S1 and S2). The sample is a high-quality 300 nm thick highly oriented V₂O₃ film (3 nm RMS surface roughness; see SI and fig. S8) grown on sapphire (Methods) that displays a five-orders of magnitude increase in electrical resistance across the IMT from 180K to 150K (Fig. 1d) with a cooling/warming hysteresis of 6K, characteristic of a first-order phase transition. We recorded nano-IR images upon cooling from room temperature across the IMT down to *T*=24 K with a temperature interval of 2-4K in the thermal window 160K - 180K, followed by a similar number of measurements upon warming to room temperature. All measurements were conducted in an ultra-high vacuum (<10⁻⁸ mbar) environment to prevent surface contamination. Lithographically patterned gold pad electrodes on the film enabled *in situ* resistance measurements for accurate thermometry calibrations between nano-IR imaging and *ex situ* measurements of the same film (SI and fig. S6).

91 Throughout this work we examine images of nano-IR signal S normalized to
92 an absolute reference through inclusion of a gold electrode within the field of view
93 (FOV) (not shown; see SI and figs. S3-5 for details of nano-IR image preparation).
94 Nano-IR signals are a capable probe of local metallicity and therefore of the nano-
95 scale IMT in correlated oxides^{22,14,20}. Metallic regions where the DC -conductivity is
96 high and the real part of the dielectric function is negative (at the probing IR
97 frequency) yield high nano-IR signals comparable to that of good metals (*viz.* gold).
98 Here, we apply a color scheme in which *red* represents high nano-IR signal,
99 indicating metallic regions. In contrast, insulating domains where the dielectric
100 function is positive produce significantly reduced nano-IR signal^{22,20} and are
101 represented in *blue* within our color scheme. Fig. 1c presents a subset of images
102 extracted from a spatially constant $20 \times 20 \mu\text{m}^2$ FOV at select temperatures through
103 a cooling and warming cycle (acquisition details in SI) measured at 920 cm^{-1} (~ 11
104 μm wavelength) well within the free-carrier (Drude) optical response of V_2O_3
105 resolved by far-field spectroscopy^{12,10,9}. The most prominent features of these
106 images are bi-directional stripes emerging through the midst of the transition. Upon
107 cooling from the metallic state (204 K), stripes of insulating material appear (175
108 K), grow (172 K), and fragment the metallic state (169 K), producing a striped
109 pattern of metallicity. At temperatures associated with rapid growth in film
110 resistance (160-170 K), metallic stripes disconnect (164 K), disorder into droplets,
111 and subsequently vanish into an insulating background (162 K). The transition
112 follows a reverse trajectory upon heating, albeit with a 6 K hysteresis. Images

acquired at higher spatial resolution (25 nm pixels) are presented in Fig. 1b, clearly showing the growth of metallic domains in an insulating background upon warming.

We draw several qualitative conclusions from our images. First, the IMT progresses upon decreasing temperature through four successive stages: i) a homogeneous metallic state, ii) a stripe-like nano-texture of percolating electronic phase coexistence, iii) an inhomogeneous insulating state supporting persistent metallic droplets, and iv) a final homogenous correlated insulating state. Second, the bipartite character of phase coexistence suggests a first-order phase transition, in accord with conventional expectations for the discontinuous Mott transition into a magnetically ordered state (AFI)^{9,8,23}. The majority of “domain walls” between metallic and insulating regions are sharp within our spatial resolution (Fig. 1b). Third, nano-IR signal levels among the distinct phases evolve gradually with temperature across the transition uncovering *bona fide* thermal evolution of the electronic response, in contrast to a putative “monolithic” phase transition between static end-phases. Fig. 2a quantifies the *dynamically binary* character of the transition through a histogram representation of nano-IR signals recorded at distinct temperatures upon warming. Similar to the case of VO₂²⁴, we observe that each bimodal nano-IR signal distribution comprises two distinct populations amidst the IMT. We identify these with “insulating” (dashed blue) and “metallic” (dashed red) populations, each well fit by an asymmetric (skew) normal distribution.

Whereas a possible admixture of phases below our spatial resolution cannot be ruled out, the marked temperature dependence of the observed “metallic phase”

remains consistent with continuous evolution in optical conductivity among distinct and locally homogeneous phases, attributable to a suppression of quasiparticle weight or pseudogap across the IMT as predicted theoretically²⁵ and suggested by area-averaged spectroscopies^{23,11,10}. Meanwhile, substantial temperature dependence among “insulating” regions also invites speculation. Calculations by cellular dynamical mean-field theory have proposed a narrow- to large-gap progression of the Mott insulating state in the phase-coexistent regime of the Hubbard model.²⁶ Likewise, mechanistic studies of near-field microscopy have confirmed a notably non-trivial relation between optical permittivity and nano-optical contrast²⁷, whereby a high-permittivity or narrow-gap semiconductor could present a high nano-IR signal (normalized S of order 0.5) consistent with our observations among these non-metallic regions for $T \gtrsim 170K$.

A detailed comparison with macroscopic resistivity bolsters our assignment of insulating and metallic regions: at each temperature, the point of intersection for the two normally distributed populations (Fig. 2a) yields a threshold nano-IR signal discerning metal from insulator in a most probable sense. We apply this temperature-dependent threshold S_{thresh} to assign individual pixels to populations of higher or lower conductivity (*viz.* metal or insulator). Thus binarized, we identified unambiguous individual electronic clusters and overall electronic phase fractions from our images. The dash-dotted curves of Fig. 2b obtained by this analysis display the area A_{max} (percentage of the FOV) of the largest connected *embedded-phase* cluster. Fig. 2b shows that A_{max} peaks at a temperature identifiable with the percolation threshold T_{perc} . At this temperature the embedding phase switches in

identity from “metallic” to “insulating” upon cooling, or *vice versa* upon warming. T_{perc} thus marks the percolation event, *i.e.* the formation of an “infinite” conducting pathway across the entire FOV and, we infer, the whole V_2O_3 film. Meanwhile, T_{perc} is also identifiable from the film’s temperature dependent electrical resistance R and conductance $G = R^{-1}$. When viewed as an effective circuit comprising parallel conductive pathways, the greatest change in film conductance is expected when the largest conductive pathway is formed or removed. Accordingly, the rate of change in film conductance with respect to temperature $\partial G \equiv \frac{d}{dT} R^{-1}$ (solid curves in Fig. 2b, obtained from *ex situ* resistance measurements) reaches a maximum value when the largest conducting pathway (spanning the film electrodes) is broken up. The comparison between A_{max} and ∂G (Fig. 2b) shows that percolation thresholds identified both *in* and *ex situ* agree to within 1 K and percolation occurs at 168 K (174 K) upon cooling (warming).

Having substantiated a nano-scale classification of metal and insulator, our binary images therefore estimate relative areal fractions of insulating and metallic phases. Moreover, *ex situ* XRD measurements of the same V_2O_3 film reveal relative intensities of corundum and monoclinic diffraction peaks (Methods), allowing assessment of constituent structural phase fractions (Fig. 2c). Fig. 2d compares the thermometry-calibrated (see SI) metallic fraction (symbols) obtained by nano-IR to the corundum fraction (solid curves) obtained by XRD, revealing a surprising 6K thermal offset between electronic and structural transitions. Consistent with the percolation temperature identified *in* and *ex situ*, we find the metallic constituent is about 60% at 168 K while cooling, whereas the occupation of the corundum phase

at this temperature remains $\sim 25\%$, with an uncertainty of about 10%. This strongly implies that some metallic regions exhibit the monoclinic structure. Correlative nano-IR and XRD measurements thus provide the first strong evidence for asynchronous electronic and structural transitions in a V_2O_3 film.

The distribution of insulator-metal transition temperatures (T_{IMT}) obtained on the basis of our binary analysis is presented in Fig. 3a, with data from cooling and warming consolidated together (after removing the relative 6K hysteresis). We identify the marked peak of the distribution with a characteristic temperature T_E for the electronic transition, equal to 164K (169K) for cooling (warming). The IMT occurs with greatest spatial prevalence at this temperature, a full 10K below the structural phase transition temperature T_{SPT} , where the SPT occurs most rapidly (and where structural phases are observed at approximately 50% fraction). Fig. 3b presents a phase diagram comprised of relevant phase fractions (PF) obtained from our data, with electronic and structural transitions shown on orthogonal axes. Most importantly, the temperature range for appearance of putative monoclinic metal (MM) is denoted in proximity to T_E .

The most striking feature initially resolved by our images with decreasing temperature is the formation of bi-directional stripes, suggesting an underlying organizational principle that could further elucidate interplay between structural and electronic transitions. To characterize growth of these stripes, we measure the extent of orientational anisotropy presented by domain walls – the boundaries between metal and insulator in our binarized images – as a function of temperature

(SI and Fig. S7). Fig. 3c shows that domain wall anisotropy is maximized 10K above T_E , matching well the structural phase transition temperature T_{SPT} . Likewise, we detect a pattern of topographic corrugations (Fig. 3e) emerging with the underlying SPT (SI and fig. S8). Nanometer-scale surface buckling follows from the differential unit cell volumes of coexisting structural domains^{28,14} and its topographic detection by our AFM broadly coincides in real-space with the striped electronic nano-texture (Fig. 3d and fig. S9). By way of this buckling, irreversible damage to the film is avoided amidst the SPT through some out-of-plane relief of accommodation strain. The buckling pattern spatially correlates with our nano-IR images most strongly at temperatures where the IMT and SPT overlap (SI and fig. S9). Moreover, this striped nano-texture also exhibits periodicity that is best revealed through the *static structure factor*, obtained as the spatial Fourier transform of the 2-dimensional correlation function for phase coexistence, which we compute for each nano-IR image (Methods)^{6,29}. Panels in Fig. 3f present the structure factor amplitude at three characteristic cooling temperatures: close to T_{SPT} (T_E+10K), above T_E (T_E+4K), and below T_E (T_E-5K). Peaks in the structure factor are most distinct near T_{SPT} . These quantify preferential wave-vectors for the formation of stripes, here spaced at 60° or 120° angular separations, explicitly revealing that 3-fold rotational symmetry of the high-temperature corundum structure is broken by at least two monoclinic twin configurations³⁰. A third expected crystallographic twin is unobserved in our images, likely due to elastic mismatch of this monoclinic twin with the sapphire substrate.

Meanwhile, the average length scale associated with these wave-vectors
 conveys the natural periodicity associated with phase coexistence in our film^{31,32},
 which we denote the structural correlation length ξ_{struct} . Fig. 4a presents a graphical
 definition of ξ_{struct} , superimposed on one section of the correlation function obtained
 perpendicular to stripes observed near T_{SPT} (fig. S10). As shown in Fig. 4b, ξ_{struct}
 rises abruptly to a value of about 1.2 μm with a broad plateau centered also in the
 temperature range identified by XRD with the SPT. The association of long-range
 spatial correlations and striped texture nano-texture with T_{SPT} is unmistakable.
 Indeed, a well-known real-space pattern of equilibrium structural phase coexistence
 (SPC) can emerge spontaneously during a SPT, taking the form of a striped “tweed”
 texture (Fig. 4c) to minimize elastic strain energies among structural domains and
 the substrate^{33,31,32,34}. Detailed studies of the sister compound VO_2 have revealed
 similar real-space patterns of SPC tunable by temperature and by intrinsic or
 extrinsic stress^{35,28,36,37}. Therefore, observed temperature dependent anisotropy
 and ξ_{struct} both suggest that electronic nano-texture observed in our images within
 this stage of the transition is largely a consequence of the IMT “guided” by the
 underlying SPT. Within this $\sim 20\text{K}$ temperature window, lattice mismatch between
 monoclinic and corundum domains results in long-range accommodation strain
 whose real-space patterns imprint on the local electronic phase.

At the final stages of the transition, we meanwhile observe clues concerning
 the character of low-temperature metallicity in our V_2O_3 film. The
 phenomenological Landau theory of phase transitions holds that a thermally
 fluctuating balance between volumetric and surface free energies among competing

phases (attributable in the Mott transition to electronic degrees of freedom²⁵)
 dictates the spatial extent of short-range correlations³⁸. We denote this spatial scale
 the electronic correlation length ξ_{elec} , and extract it at each temperature from the
 central full-width at half maximum of the correlation function (Fig. 4a; see
 Methods). Shown in Fig. 4e and 4f, ξ_{elec} quantifies the characteristic length scale for
 locally correlated electronic domains, reaching a maximum (~ 400 nm) at T_E . The
 growth and peak in ξ_{elec} reveals the tendency for metallic domains to form
 “droplets” near T_E rather than stripes (Figs. 4f and 4i), and resembles the strong
 temperature dependence of spatial correlations more characteristic of continuous
 phase transitions, which can exhibit universal scaling near criticality^{38,17}. Whereas
 influence of the STP complicates unambiguous quantitative extraction of ξ_{elec} versus
 T , throughout the transition we nevertheless observe robust critical scaling in the
 temperature dependence of the largest electronic domain size $d_{\text{max}} \equiv \sqrt{A_{\text{max}}}$,
 observed to scale as $|T - T_{\text{perc}}|^{-\nu}$ with $\nu = 0.96 \pm 0.07 \approx 1$ (Fig. 4d). Noting that
 metallic percolation extends predominantly from the largest metallic cluster, our
 directly resolved scaling shows quantitative agreement with the scale invariant
 nucleation site density inferred from area-averaged optical studies of V_2O_3 films
 grown by the same method (Methods)³⁹.

Although we cannot rule out an origin for these critical behaviors prosaically
 tied to disorder (*e.g.* the Anderson-Mott transition), it is noteworthy that our V_2O_3
 film presents a metal-insulator transition among the cleanest (most abrupt) yet
 reported. In the clean limit, whereas critical scaling behaviors are conventionally at
 odds with a bulk first order transition, they might here be reconciled with

predictions for phase separation in the Hubbard model. Below the Mott transition temperature, uniform band filling in the homogeneous state is proposed to become unstable at fixed chemical potential μ , and the system phase-separates into regions with distinct charge density n associated with the insulating state at half band-filling and a slightly doped metallic state.⁴⁰ This charge disproportionation Δn is theorized to reach a few percent upon decreasing temperature. Consequently, long-ranged Coulomb interactions are expected to penalize macroscopic charge imbalance, whereas positive surface tension favors few coexistent domains, resulting in glassy electronic phase coexistence with droplets of characteristic size $\xi_{\text{elec}} \sim (\frac{\sigma}{\Delta n^2})^{1/d}$ in a d -dimensional IMT^{41,42}. Tied to growing Δn for temperatures below T_{IMT} ⁴⁰, *Coulomb frustration* intensifying away from the local transition temperature could underlie our observation of metallic droplets persisting at rapidly suppressed length scales amidst this IMT (Figs. 4d and e). We speculate the behavior of $\xi_{\text{elec}}(T)$ might provide spatial sensitivity to $\Delta n(T)$ at experimentally fixed chemical potential, a real-space window into the μ - n phase diagram for coexistent phases of the transition.

We can confirm directly that the IMT proceeds by a first-order electronic phase transition even among these low temperature metallic droplets. Enabled by colocalized nano-imaging within our FOV, Fig. 4g presents characteristic transition curves (σ vs. T) for *loci* of pixels that exhibit the same transition temperature T_{IMT} upon cooling (associated spatial maps of T_{IMT} presented in fig. S11). Taking the initial transition curve at $T_{\text{IMT}}=176\text{K}$ for reference, Fig. 4h reveals a sharp first-order discontinuity in σ for all T_{IMT} , and by implication, abrupt decrease in the free carrier

optical response. Sir Neville Mott's idealization of this IMT predicted such discontinuity on the basis of the long-range Coulomb interaction⁸. Even without lattice coupling, studies of the Hubbard model including beyond nearest-neighbor interactions have postulated a first-order Mott transition^{43,9,40}, consistent with our observations among metallic droplets, upon cooling many of which have presumably already undergone the structural transition.

Our observation of this low temperature metallic phase (Fig. 4i) aligns with reports for the related correlated oxide VO₂^{15,22,16,14,29,44}. Whereas a transient or pressure-induced MM has been observed even in VO₂ single-crystals^{45,46}, its more robust appearance in extrinsically or epitaxially strained samples suggests a general mechanism decoupling the IMT from the SPT. Studies of the semi-infinite Hubbard model for example have predicted stable interfacial metallicity amidst a bulk Mott insulating state⁴⁷. Meanwhile, X-ray absorption studies of V₂O₃ have identified inequivalent metallic states attained through purely thermal or pressure-driven transitions⁴⁸ and have revealed a novel pressure-induced MM phase¹³, perhaps related to that detected here.

We propose that epitaxial strain and the consequent striped SPT nanotexture may play a crucial role among films. Stabilization of an intermediate monoclinic metal may demand a strain environment fostered by pervasive structural domain walls, implicit within the striped nanotexture we resolve in this correlated insulator. Indeed, a stabilized intermediate electronic state with attributes of the high-temperature phase has already been observed at temperatures below the

magnetoresistive transition of manganite films, attributed likewise to the accommodation strain of coexistent structural phases.⁴⁹ Consequently, we speculate that a monoclinic metal may assist in strain relief at structural domain boundaries. Moreover, growing Coulomb frustration below T_{IMT} may afford further stability to an intermediary phase in our V_2O_3 film.

Associating the monoclinic metal with a strain-induced or Coulomb-frustrated phase suggests novel real-space facets of the electronic (Mott) transition in this correlated insulator, hitherto unexplored at this spatial resolution. Our study reveals the rich impact of nanotextured inhomogeneity on the low temperature insulator-metal transition in V_2O_3 , affirming that exotically rich behaviors can underlie even this classic insulator-metal transition, moreover demanding amendments to the bulk V_2O_3 phase diagram for epitaxial structures^{17,8}. Low-temperature nano-spectroscopic probes and X-ray magnetic imaging will be imperative to further elucidate the electronic character of exotic phases emerging amidst the insulator-metal transition in V_2O_3 and other correlated oxides.

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Author Contributions:

340 S. Wang and J. G. Ramirez grew the V_2O_3 film. A. S. McLeod and E. van
341 Heumen performed nano-IR measurements on the film, whereas J. G. Ramirez
342 and T. Saerbeck performed x-ray diffraction and *ex situ* resistance
343 measurements on the film. A. S. McLeod, E. van Heumen, M. Goldflam, L.
344 Anderegg, and P. Kelly developed the nano-IR instrumentation. All authors
345 prepared the manuscript.

Methods

V₂O₃ Films

The V₂O₃ film studied in this work was epitaxially grown on (012)-plane sapphire substrate by RF magnetron sputtering from a V₂O₃ target (1.5" diameter, > 99.7%, ACI Alloys, Inc.). The samples are prepared in a high vacuum system with a base pressure of 1×10^{-7} Torr. The substrate temperature was kept at 750°C during the deposition. 4 mTorr ultrahigh purity (99.999%) Ar and 100 W RF power were used for the deposition of V₂O₃. These conditions yield a deposition rate of 0.67 Å/s. Based on X-ray diffraction characterization of the film, compressive strain from lattice mismatch between hexagonal lattice constants for room temperature sapphire and V₂O₃, combined with stress relief by film buckling and granularity, induces lattice expansion along the film c-axis and a concomitant increase in the c/a ratio relative to bulk crystals. Previous studies of sapphire-grown V₂O₃ films have equated this expansion with tensile hydrostatic chemical pressure attainable through chromium doping, affecting a putative decrease in bandwidth and increase in the Mott transition temperature⁵⁰. Indeed, the transition temperature for our film is as much as 16K higher than for bulk crystalline V₂O₃. Notably, previous similarly grown films displayed characteristics similar to the best available single crystal samples.

Determination of structural phase fractions by X-ray diffraction

We have determined the SPT temperature and the crystallographic phase fraction in our film by performing temperature-dependent X-ray diffraction (XRD)

measurements. We measured the out-of-plane XRD of the rhombohedral (012) peak shifts from $2\theta = 24.30^\circ$ above the SPT (300 K) to the monoclinic (011) $2\theta = 24.05^\circ$ below it (100 K). The peaks were then fitted with two Gaussian curves; keeping fixed the 2θ values of the low and high temperature phases. The area under each Gaussian was normalized to the total area and the percentage volume fraction of each phase was thus obtained. The SPT temperature is obtained at the temperature at which both phases are equally populated.

Correlation Analysis

Informally, the correlation function $g(\delta\vec{r})$ of an image $I(\vec{r})$ (evaluated at lateral positions $\vec{r}$) expresses the level of statistical similarity between image features separated by a displacement $\delta\vec{r}$. The correlation function is formally given by³⁸:

$$g(\delta\vec{r}) = \langle \delta I(\vec{r}), \delta I(\vec{r} + \delta\vec{r}) \rangle = \int d^2\vec{r} [I(\vec{r}) - \langle I(\vec{r}) \rangle] [I(\vec{r} + \delta\vec{r}) - \langle I(\vec{r} + \delta\vec{r}) \rangle]. \quad (1)$$

Here $\langle \dots \rangle$ indicates an areal average of the enclosed value. Appearance of the mean value $\langle I(\vec{r}) \rangle$ in Eq. (1) reflects that only image inhomogeneities (“features”) are relevant for the characterization of spatial correlations. Eq. (1) was used to compute the correlation function for each nano-IR image across the insulator-metal transition of our V_2O_3 film. Each correlation function was normalized to unity at $\delta\vec{r} = \vec{0}$, taken by construction to indicate 100% correlation.

The correlation function is known to reveal intrinsic periodicities more clearly within noisy data than would be possible to identify through direct inspection alone. We leverage this feature to identify periodicity in our nano-IR images, whose mean

length scale we identify as ξ_{struct} . Meanwhile, the rotational average of the correlation function (yielding $g(r)$, evaluated at displacement magnitude r alone) was used to identify ξ_{elec} according to its central full-width at half-maximum. This correlation length reflects the typical scale for statistical correlations to locally decay by e^{-1} and corresponds intuitively with the average dimension of characteristic image inhomogeneities. In our case these comprise droplets of the minority electronic phase – whether insulating in character above the percolation temperature, or metallic below. The “divergent” character of ξ_{elec} revealed by our correlation analysis and the thermal scaling of largest electronic cluster sizes both qualitatively match the phenomenology of Landau theory for scale-invariant spatial fluctuations proximate to a *continuous* phase transition, or alternatively a scenario of temperature-dependent Coulomb frustration.^{42,41,40}

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Figure 1 | nano-IR imaging of the low temperature Mott transition in V_2O_3 . a) Schematic depiction of near-field microscopy of phase coexistence in a V_2O_3 thin film; nano-IR signal S superimposed on film topography at 171K (cooling). b) High-resolution co-localized near-field images of coexisting phases evolving upon warming the film from the antiferromagnetic insulator (AFI) to paramagnetic metal (PM) phase; 1 micron scale bar; color scale as in c). c) Large-area co-localized nano-IR images of the electronic phase transition upon cooling (blue arrows) and warming (orange arrows); 5 microns scale bar. The color scale (bottom) distinguishes metallic from insulating regions. d) Resistance of the film versus

415 temperature upon cooling (blue arrow) and warming (orange arrow); dashed lines
416 demarcate the temperature range of phase-coexistence in the insulator-to-metal
417 transition (IMT). Inset: schematic arrangement of gold pads on the film surface
418 used as in-situ transport electrodes and for quantitative normalization of near-field
419 images.

Figure 2 | Binary analysis of phase populations. a) Histogram representation of nano-IR signals (*viz.* levels of free carrier optical response) recorded from the V_2O_3 film at several temperatures upon warming from 164K to 184K relative to an ideal metal (gold). An example fit to the distribution at 173K by two asymmetric normal distributions reveals insulating (dashed blue) and metallic (dashed red) populations evolving with temperature and demarcated by a threshold nano-IR signal level (S_{thresh}); note vertical log scale. b) Comparison of the largest electronic cluster size observed by nano-IR imaging (A_{max}) against changes in film transport conductance per unit temperature ∂G for both cooling and warming; both metrics identify a coincident percolation temperature T_{perc} . c) Bimodal decomposition of diffraction peaks measured by X-ray diffraction (XRD) admits measurement of structural phase fractions (Methods). d) Thermometry-calibrated comparison of phase fractions for

434 the metallic phase identified by nano-IR imaging against those for the high-
435 temperature structural phase (corundum) identified by XRD; an anomalous thermal
436 offset suggests persistent metallicity in the monoclinic structure.

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440 **Figure 3 | Phase diagram of electronic and structural phases in thin-film V_2O_3 .**

441 a) Distribution of insulator-metal transition temperatures T_{IMT} obtained as the
 442 fraction of observed pixels to transition per degree K; data from cooling and
 443 warming together. The peak in the transition indicates the most likely transition
 444 temperature, denoted T_E . b) Phase diagram presenting T -dependent fill fractions
 445 (FF) for electronic and structural phases (orthogonal axes) measured by nano-IR
 446 imaging and X-ray diffraction (XRD). *EPC* = electronic phase coexistence; *SPT* =
 447 structural phase coexistence; *MM* = monoclinic metal. c) Directional anisotropy of
 448 electronic domain walls versus temperature as obtained from a binary insulator-
 449 metal classification of nano-IR image pixels; anisotropy maximizes abruptly at the
 450 structural phase transition temperature T_{SPT} , associating electronic anisotropy with
 451 "guiding" by the SPT. d) Subset of nano-IR image at $T_E + 4K$ (warming) compared

452 with e) relative topographic corrugations detected simultaneously by AFM. f) Static
453 structure factor (see text) of EPC measured near T_{SPT} , above T_{E} , and below T_{E} ,
454 revealing preferred structural wave-vectors emerging at 60° lateral separations;
455 temperatures indicated by dots on the phase diagram panel b).

Figure 4 | Characteristics of persistent metallicity. a) Trace from the image correlation function obtained 9K above T_E , perpendicular to the direction of stripes; relevant correlation lengths are indicated. b) The structural correlation length associated with periodic correlations; ξ_{struct} plateaus at T_{SPT} and originates from structural phase coexistence. c,f,i) Acquisition temperatures for nano-IR images indicated by colored dots in panel e). c) Indication of ξ_{struct} associated with real-space stripe periodicity; scale bar: 2 microns. d) Scale invariance of the largest electronic domain size d_{max} , fitting a power law with critical exponent $\nu \approx 1$ close to the percolation threshold temperature T_{perc} . e) The electronic correlation length associated with short-range correlations; ξ_{elec} peaks abruptly at T_E . f) Indication of

468 ξ_{elec} associated with the characteristic size of metallic droplets; field of view is a
469 subset of panel c); scale bar: 1 micron. g) Average transition curves (nano-IR signal
470 σ vs. T) acquired from pixels exhibiting insulator-metal transition temperatures
471 $T_{\text{IMT}}=176\text{K}$ to 160K upon cooling. h) Curves from g) after referencing to $T_{\text{IMT}}=176\text{K}$,
472 revealing a first-order discontinuity in σ down to $T_{\text{IMT}}=160\text{K}$. i) Putative monoclinic
473 metallic matches persisting even 10K below T_E , and further yet below T_{SPT} ; scale
474 bar: 1 micron.