Nanoscale dynamics across the Mott transition in V$_2$O$_3$

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The phase diagrams of 3$d$ metal oxides provide rich landscapes to explore the non-equilibrium degrees of freedoms during an insulator-to-metal transition (IMT). In these materials, the dynamics of nano-textured insulating and metallic phases is characterized by an unexplored complexity than enables manipulation of phase separation to control the properties of quantum materials on ultrafast timescales. Here, we combine X-ray photoemission electron microscopy and non-equilibrium optical spectroscopy to link the temporal and spatial dynamics of the IMT in the Mott insulator V$_2$O$_3$. We show that metallic droplets, which form at the boundaries of striped insulating domains, act as seeds for the non-equilibrium expansion of the metallic phase triggered by the photo-induced change in the 3$d$-orbital occupation. We demonstrate that the growth of the metallic phase can be controlled by properly tailoring the light-excitation protocol. Our results unveil the coupled electronic and structural dynamics during an ultrafast IMT and open up the possibility of controlling the ultrafast dynamics of Mott transitions in a way that is inaccessible by thermal means.

Insulator-to-metal transitions (IMT) in strongly correlated materials are among the most remarkable examples of first-order solid-solid transformations driven by electronic interactions [1]. The transition from the Mott insulating state, in which the Coulomb repulsion suppresses the motion of charge carriers throughout the lattice, to the metallic phase has been the subject of intense interdisciplinary studies. While ultracold atoms trapped in "ideal" optical lattices have been used to investigate the dynamics of the Mott IMT in the absence of coupling to any external bath [2,3], the solid-state counterpart is much more complex as a consequence of the inherent interplay between the electronic, lattice and spin degrees of freedom [1]. Furthermore, the intrinsic nano-texture of the IMT confines the transition dynamics to the picosecond timescale [12,13], which is inaccessible by conventional dynamical studies. Addressing metallic domain nucleation and growth therefore provides fundamental insights on the forces and interactions driving the phase transformation and is key to controlling IMT at THz frequencies [14,15].

Here, we combine element-specific X-ray nanoimaging with non-equilibrium optical spectroscopy to investigate the prototypical Mott insulator V$_2$O$_3$ [16], which undergoes a first-order IMT from an antiferromagnetic monoclinic insulator to a paramagnetic rhombohedral metal. We unveil the existence of striped insulating domains, whose boundaries seed the formation of metallic nuclei within the insulator-metal coexistence region. We also establish the link between this intrinsic nano-texture and the dynamics triggered by impulsive light excitation, which induces a sudden change of the free energy density difference, $\delta g = \delta (g_M - g_I)$, between the two phases ($g_I$: insulating, $g_M$: metallic). We follow the ballistic growth of metallic nuclei within the insulating domains and we demonstrate the possibility of using light to control their dynamics.

The low-energy physics of vanadium sesquioxide (V$_2$O$_3$) originates from the interactions between the two electrons occupying the V-$3d$ levels and from their coupling to the lattice deformations. In particular, the octahedral crystal-field splits the V-$3d$ orbitals into lower $t_{2g}$ and upper $e_g$ bands. Since the octahedra display a further trigonal distortion, reinforced by the on-site Coulomb repulsion ($U \approx 2.5$ eV [17]), the $t_{2g}$ orbitals are split (see Fig. 1) in turn into a lower $e_g^T$ doublet, mainly oriented in the $a$-$b$ plane, and an upper $a_{1g}$ singlet, with lobes oriented along the $c$-axis. In the low-$T$ antiferromagnetic insulating (AFI) monoclinic phase, the average occupancy of $e_g^T$ orbitals ($n_{e_g^T} \approx 0.83$) largely overcomes that of the $a_{1g}$ level ($n_{a_{1g}} \approx 0.17$) and the system behaves as a half-filled two-band Mott insulator [18] with an effective gap $2\Delta_{\text{eff}} \approx 0.5$ eV. At $T_{\text{IMT}} \approx 160$ K, the structural change from monoclinic to corundum accompanies a jump of the $a_{1g}$ occupation with a concomitant transition to the paramagnetic metallic (PM) phase.
FIG. 1. Nanoscale insulating domains imaged by XMLD-PEEM. a) Left: X-ray Absorption Spectroscopy (XAS) spectrum showing the $L_{2,3}$ vanadium absorption lines. The light blue areas highlight the spectral regions where the maximum XMLD signal is obtained. Middle: Sketch of the $L_{2,3}$ transitions from the V-2p levels to the doubly occupied V-$t_{2g}$ levels further split into $a_{1g}$ and $e^{\pi}_{g}$ by the trigonal distortion of the lattice. Right: trigonal distortion of the V atoms along the c-axis of the hexagonal unit cell. b) Left: a-b plane projection of the monoclinic lattice structure in the low-T insulating phase. Right: a-b plane projection of the corundum lattice structure in the high-T PM phase. c) Left top: XMLD-PEEM image taken at 120 K (heating branch of the hysteresis) evidencing striped AFI domains. Left bottom: XMLD intensity distribution (thick grey line) of the image. The analysis evidences a broad zero-centered distribution (green), corresponding to the resolution-broadened edges of the striped domains, and three distinct distributions with non-zero signal (red, blue, light blue), reflecting the different structural nanodomains within the AFI phase. Middle: hysteresis cycle as obtained by resistivity measurements and by the metallic filling analysis of the temperature-dependent XMLD-PEEM images (see Methods). The yellow (green) arrows indicate the heating (cooling) cycles. Right top: XMLD-PEEM image taken at 200 K (heating branch of the hysteresis) showing a homogeneous background, typical of the PM metallic phase. The residual red spots are due to defects used as reference to precisely align the XMLD-PEEM images. Right bottom: XMLD intensity distribution (thick grey line) of the image. The analysis evidences a zero-centered single-mode distribution.
characterized by $n_{a_{1g}} \simeq 0.25$ [13]. Across the IMT, the electrical resistivity can be tuned over several orders of magnitude by means of small variations in chemical composition [19–21], pressure [22–25], temperature [10] and, more recently, by external application of electric fields [26–28] or irradiation with short light pulses [12, 29–32]. Snapshots of the IMT in real space have been taken via photoemission electron microscopy (PEEM) on a 40 nm V$_2$O$_3$ film epitaxially grown on a (0001)-Al$_2$O$_3$ substrate, therefore with the c-axis oriented perpendicular to the surface of the film (see Methods). PEEM images have been obtained by exploiting X-ray Magnetic Linear Dichroism (XMLD) [33] at the V-L$_{2,3}$-edge (∼513-530 eV) as the contrast mechanism. In the AFI phase, the XMLD signal at the specific energies of 513 and 520 eV arises from the absorption difference between X-rays polarized parallel and perpendicular to the $a_{1g}$ orbitals [18], which have lobes oriented along the c-axis. The different contrast between the AFI and PM phases originates from the different occupancy ratio, $n_{a_{1g}}/n_{e_{g}}$, within the V-L$_{2g}$ orbitals [15, 34]. In Fig. 1 (left) we report the spatially-resolved XMLD contrast in the AFI phase ($T=120$ K). Striped insulating nano-domains, oriented along the hexagonal crystallographic axes, are clearly visible with characteristic dimensions of a few micrometers in length and 200-400 nm in width. The analysis of the intensity distribution (see Methods) highlights the presence of three different domains, which correspond to three possible orientations of the monoclinic domains subject to distortions along the three equivalent edges of the hexagonal unit cell [15, 35]. The XMLD contrast depends [36] on the relative orientation between the almost in-plane antiferromagnetic axis within each domain and the axis defined by the electric-field vector of the linearly polarized X-rays and is controlled by rotating the sample around the c-axis (see Supplementary Information). We point out that, under temperature cycling, the striped domains always nucleate at the same location (see Supplementary Information), thus suggesting that the film-substrate interaction drives the minimization of the elastic energy associated with the volume increase of the AFI phase [13]. When the temperature is increased, the XMLD contrast of the insulating domains progressively vanishes until the uniform metallic phase, characterized by a single-color distribution, emerges (see right-top panel in Fig. 1). The metallic filling $F_M$ is calculated as the ratio between the number of metallic pixels (green XMLD contrast), recognized by pixel-based segmentation techniques (see Methods), and the total number of pixels within a fixed insulating domain. The temperature dependence of $F_M$ is shown in the bottom-middle panel of Fig. 1. The metallic filling displays a large temperature hysteresis ($\delta T \sim 20$ K), which corresponds to the resistivity hysteresis measured on the same sample and reported in the top-middle panel of Fig. 1.

The complex intrinsic nano-texture of the insulating phase constitutes the template for metallic nuclei formation and growth, thus leading to the global transformation from the AFI to the PM phase. The temporal dynamics of this transition was investigated by time-resolved optical spectroscopy, which employs ultrashort light pulses to impulsively weaken the AFI phase and drive the phase transformation. This process can be understood starting from the following thermodynamic considerations. At the equilibrium temperature $T$, the distribution function of stable metallic nuclei, i.e. those with volume of the order of the critical volume $v_{cr}$, is $f_{eq}(v_{cr}) \propto \exp(-G_{cr}/kT)$, where $G_{cr}$, is the energy barrier necessary to form a stable nucleus. This energy barrier, $G_{cr}=(g_M-g_I)v_{cr}+\alpha \kappa_{cr}$, is a balance between the volume energy gain to form metallic nuclei ($g_M < g_I$) and the surface energy loss associated with the surface tension ($\alpha$) [37, 38] and critical surface area ($\kappa_{cr}$). The effect of the excitation with light pulses shorter (∼50 fs) than the typical dynamics of the phase transformation and with photon energy ($\hbar \omega=1.55$ eV) larger than the charge-gap is thus twofold. On one hand, the energy absorbed is rapidly released to the lattice and leads to a quick increase ($\delta T$) of the local effective temperature within less than one picosecond [29–32]. On the other hand, the impulsive photoexcitation modifies the population of the $e_{g}$ and $a_{1g}$ orbitals, which constitutes the control parameter of the IMT [32, 39, 40]. The light thus induces a change in the orbital polarization, $\delta p = \delta(n_{a_{1g}}-n_{e_{g}})$, that weakens the Mott insulating phase and drives a further decrease of the free energy density difference between the

**FIG. 2. Energy-resolved differential reflectivity variation.** a) Cartoon of the photoexcitation across the Mott gap. The change in the occupation of the $a_{1g}$ and $e_{g}$ orbitals gives rise to the orbital polarization change $\delta p = \delta(n_{a_{1g}}-n_{e_{g}})$, which decreases the energy difference between the $a_{1g}$ and $e_{g}$ levels. The photoinduced redshift of the effective gap, indicated by the black arrow, corresponds to a weakening of the insulating state and thus triggers the growth of metallic nuclei. b) Long-time reflectivity variation, $\delta R/R(\omega, 25$ ps), as a function of the probe energy, $\hbar \omega$, at two different temperatures. The dashed lines represent the differential reflectivity modeled by assuming a redshift $\delta \omega \sim 4$ meV of the Lorentz oscillator representing the first interband transition at $h \omega \approx 1.17$ eV ($T=140$ K) and $h \omega \approx 1.24$ eV ($T=160$ K). The green area represents the optical conductivity (arbitrary units) associated with the optical transition at $\sim 1.2$ eV. The green arrow represents the photoinduced redshift of the interband transition.
insulating and metallic phases, as given by [39]:

$$\delta(g_M - g_I) \sim -\frac{U}{2} \delta \rho$$  \hspace{1cm} (1)

The light-induced increase of \(T\) and the simultaneous decrease of \(G_{cr}\) is the microscopic mechanism that, in the \(I-M\) coexisting region, can lead to the generation and growth of metallic nuclei within the insulating domains, up to the point of transforming the entire excited volume into a homogeneous metallic phase.

The dynamics of the photoinduced phase transformation is snapped via broadband (0.8-2 eV) pump-probe reflectivity measurements, which probe the change of the optical properties as a function of the delay \(\Delta t\) from the pump excitation. In multi-orbital Mott insulators, such as \(V_2O_3\), the photoinduced orbital polarization gives rise to a redshift of the effective charge gap given by: \(\delta \Delta_{\text{eff}} \simeq (g_M - g_I)\) [39][41], as depicted in Fig. 2. In Fig. 2b we report the measured reflectivity change, \(\delta R/R(\omega, \Delta t)\), as a function of the probe photon energy and at a fixed delay time \((\Delta t = 25\) ps). Reflectivity variations measured at 140 K and 160 K both display a sign change at \(\sim 0.9\) eV compatible with the expected redshift of the lowest optical transition present in the equilibrium dielectric function at \(\sim 1.2\) eV (see the optical conductivity in Fig. 2b) [17][12]. We point out that, while the sign and the amplitude of the measured \(\delta R/R\) is strongly frequency-dependent, its temporal dynamics has the same behavior over the entire probed energy range. As a consequence, we can assume that, independently of the probe frequency, the time-dependent reflectivity variation is proportional to \(\delta \Delta_{\text{eff}}\), which linearly depends on the metallic volume within the probed area, i.e. the metallic filling. In the following, we will thus assume that \(\delta R/R(\Delta t) \propto F_M(\Delta t)\), at any probe wavelength.

To prove the connection between the spatial evolution of metallic nuclei and the temporal dynamics of the phase transformation we performed time-resolved optical spectroscopy experiments on the same sample and in the same point of the hysteresis cycle as the XMLD-PEEM measurements. In Fig. 3a, we show the one-to-one correspondence between the real space XMLD-PEEM images of the \(IMT\) and the time-resolved dynamics during the heating cycle. When focusing on specific insulating domains (see black contours in the XMLD-PEEM images shown in the top row of Fig. 3b) we observe the emergence of finite-size metallic nuclei, i.e. regions with a critical width larger than the experimental resolution of \(30\) nm, at temperatures above \(145\) K. We note that the metallic phase nucleates along the boundaries between different insulating domains, as expected for a martensitic transition [39]. When further increasing the temperature, the growth of the elongated metallic nuclei leads to a percolative transition [10] in a narrow temperature range (155-165 K). At the end of the \(IMT\) (180 K), the metallic regions eventually occupy most of the pristine insulating domains and \(F_M \sim 1\). In the bottom row of Fig. 3b we report the dynamics of \(F_M(\Delta t)\), as measured by time-resolved optics with an incident pump fluence of \(0.3\) mJ/cm\(^2\). Assuming that all the absorbed energy \((E_{\text{abs}} \sim 13\) J/cm\(^3\)) is converted into local heat, the maximum temperature increase is limited to \(\delta T \lesssim 4\) K (see Methods), i.e. a value by far smaller than that needed to induce the \(IMT\) by thermal effects. At the same time, the light excitation induces an orbital polarization \(\delta \rho \sim 3 \times 10^{-3}\) (see Methods), corresponding to a free energy density variation \(\delta(g_M - g_I) \simeq 4\) meV, which largely overcomes the thermal effect \(k_B \delta T\). In the time-domain,
FIG. 4. **Avrami model for the metallic growth.** a) Avrami plot of the dynamics of $F_M(\Delta t)$ at different temperatures. A vertical offset has been added to the raw data. The black lines indicate the expected curves for $N=2$ and $N=3/4$. Inset: Avrami coefficient $N$ of the long-lived dynamics ($\Delta t > 1$ ps) as a function of the temperature. The red line indicates the asymptotic value $N=3/4$. b) Avrami plot of $F_M(\Delta t)$ at 145 K for different pump excitation fluences, expressed in mJ/cm$^2$. The solid lines indicate the expected curves for $N=3/4$. A vertical offset has been added to the raw data. The light-blue area highlights the temporal region in which the pump triggers the formation of metallic nuclei with $N=2$. c) Avrami plot of $F_M(\Delta t)$ at 120 K for different pump excitation fluences, expressed in mJ/cm$^2$. The black lines indicate the expected curves for $N=1/2$. The arrows indicate the time at which the maximum value of $F_M(\Delta t)$ is reached.

the existence of stable finite-size metallic regions (white contours in XMLD-PEEM images of Fig. 3a) for $T > 145$ K corresponds to a critical slowing down of the build-up dynamics of $\delta R/R(\Delta t)$, with respect to the time-trace measured at lower temperatures (120 K). In the $I$-$M$ coexisting region we also observe an increase of the long-time signal ($\Delta t \sim 25$ ps), as shown in the bottom row of Fig. 3a. In Fig. 3c we report the temperature dependence of $\delta R/R(25$ ps), which exhibits a clear maximum corresponding to the hysteresis region observed by XMLD-PEEM and shows a completely different behavior from the measured temperature-dependent metallic filling at equilibrium (black line). The physical picture emerging from the combination of nano-imaging and picosecond dynamics of the IMT is the following: in the temperature range in which the average volume of the metallic domains is smaller than $v_{cr}$, the system is dominated by the formation of fluctuating small metallic nuclei. In this regime, the light excitation drives a simple increase of the number of metallic nuclei, which are then rapidly reabsorbed by the insulating environment. In the $I$-$M$ coexisting region ($T \gtrsim 145$ K), the photo-induced orbital polarization $\delta p$ triggers the growth of the metallic nuclei already nucleated at the domain boundaries and leads to a long-lived increase of the metallic filling, which is eventually re-absorbed on the nanosecond timescale.

The growth of a new phase at the cost of the initial one is usually described by the Avrami model [43-45], which predicts $F_M(\Delta t)=1-\exp(-K\Delta t^N)$, where $K$ is a constant related to the growth rate and $N$ is the Avrami coefficient, characteristic of the phase growth mode. In Fig. 4 we plot the quantity $\ln(-\ln(1-F_M(\Delta t)))$, as a function of the logarithmic time $\ln(\Delta t)$. The normalized metallic filling is taken as the ratio between the reflectivity variation and its asymptotic value, i.e. $F_M(\Delta t)=[\delta R/R(\Delta t)]/|\delta R/R(>25$ ps)|. At very low temp-
perature ($T=80$ K, Fig. 4h), we observe a rapid (0-500 fs) growth with $N=2$, followed by a slow decay. While the fast dynamics is compatible with the sudden growth of two-dimensional domains with a rapidly exhausting nucleation rate \cite{25}, the slow relaxation to the initial equilibrium condition, i.e. $F_M=0$, indicates that the metallic nuclei never reach the stability threshold. When the temperature is increased, the dynamics of $F_M(\Delta t)$ starts exhibiting the fingerprint of a long-lived growth of the metallic phase, under the form of a linear increase of the signal, in agreement with the prediction of the Avrami law: $\ln(-\ln(1-F_M(\Delta t)))=N\ln(K)+N\ln(\Delta t)$. The Avrami coefficient progressively increases (see inset in Fig. 4a) until it reaches a plateau in the $I-M$ coexisting region ($T >145$ K), where the data show a linear behavior over more than one decade (1-20 ps). In this regime the Avrami coefficient ($N=3/4$) is very close to unity, as expected for a martensitic transition \cite{46}, in which the seeds of the new phase nucleate at the domain boundaries in the first instants and then grow with constant velocity along one direction. The growth of metallic filling saturates after $\Delta t_{sat} \sim 25-30$ ps from the impulsive excitation. Considering the sound velocity, $v_s \sim 7$ km/s \cite{17}, $\Delta t_{sat}$ corresponds to a typical length of $\sim 200$ nm, which is very close to the average width of the striped monoclinic domains observed in the AFI insulating phase. This result suggests that the metallic growth is a ballistic collective process \cite{12,13} in which the lattice distortion wavefront propagates at the sound velocity, until it encounters the edges of the striped insulating domains. In the $I-M$ coexisting region, the $N=3/4$ growth dynamics is an intrinsic characteristic, which does not depend on the pump excitation fluence, as shown in Fig. 4f, for excitations spanning the range 0.3-2.6 mJ/cm$^2$. In contrast, an interesting fluence-dependent dynamics is observed at $T=120$ K (see Fig. 4i), in close proximity of the hysteresis region. At low pump fluences, the Avrami-like growth dynamics ($N=1/2$) is limited to less than 10 ps, indicating that the ballistic increase of the corundum metallic phase stops before the metallic domains reach the edges of the insulating monoclinic stripes. When the pump fluence is increased up to 2.2 mJ/cm$^2$ ($\delta(g_M-g_I) \sim 30$ meV) the growth dynamics progressively extends to longer times and eventually reaches the maximum value of 30 ps, corresponding to the complete transformation of the insulating stripes along the transverse direction. Notably, in the high-fluence regime, the $T$-dependent Avrami coefficient (as seen in the inset of Fig. 4k) does not change with respect to the low-fluence excitation case. This result demonstrates that the increase of the time-span of the metallic growth is mainly controlled by the photoinduced orbital polarization and it is not the mere consequence of a local increase of the effective temperature after the picosecond electron-phonon thermalization.

The present experiments and results offer a new platform for the study and comprehension of the first-order Mott transition dynamics in correlated solids and pave the way to the use of light-induced orbital polarization to non-thermally control the growth dynamics of metallic domains in the vicinity of the IMT. The possibility to suitably engineer the lateral size of the nano-textured insulating domains in thin films and to use tailored light pulses to trigger and stop on demand the picosecond growth of metallicity is the key to controlling the electronic and magnetic switching of Mott insulators at THz frequencies, with impact on the development of novel Mottronics devices \cite{15}.

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**METHODS**

**A. Samples**

The 40 nm epitaxial V$_2$O$_3$ film is deposited by oxygen-assisted molecular beam epitaxy (MBE) in a vacuum chamber with a base pressure of $10^{-9}$ Torr. A (0001)-Al$_2$O$_3$ substrate is used without prior cleaning and is slowly heated to the growth temperature of 700°C, as measured with a thermo-couple. V is evaporated from an electron gun with a deposition rate of 0.1 Å/s and an oxygen partial pressure of 6.2 $\cdot$ 10$^{-6}$ Torr is used during the growth \cite{28}. Under these conditions, a single crystalline film with the c-axis oriented perpendicular to the surface is obtained (see Supplementary Information for X-ray diffraction data). Temperature dependent resistivity measurements are assessed in the Van der Pauw (VDP) configuration with Au/Cr contacts and using an Oxford Optistat CF2-V cryostat with a sweep rate of 1.5 K per minute.

**B. Photoemission electron microscopy (PEEM)**

PEEM and XAS spectra have been measured at the beamline I06 of Diamond Light Source. The spot size of the X-ray beam is 10 x 10 μm$^2$. The PEEM images
presented in this paper have been obtained by performing XMLD experiments at a photon energy of $\sim 513$ eV, in proximity of the vanadium $L_{2,3}$ edge. Each PEEM image is the average of the difference between 50 PEEM images taken with light-polarization parallel (i.e. in the $a$-$b$ plane) and perpendicular (i.e. along the $V_2O_3$ c-axis) to the film surface, normalized by their sum.

C. Time-resolved pump-probe spectroscopy

Time-resolved reflectivity measurements have been performed using three different experimental setups: i) A pump supercontinuum-probe, based on white light (from 0.8 to 3.1 eV, generated by focusing a train of ultrafast pulses (temporal width $\lesssim 1$ µJ energy/pulse in a 3 mm thick Sapphire window); ii) A pump tunable-probe setup, based on an Optical Parametric Amplifier (OPA). In both these cases, a Regenerative Amplifier (Coherent RegA 9000), generating a train of ultrafast pulses (temporal width $\sim 50$ fs) at 250 kHz with a wavelength $\lambda = 800$ nm and $\sim 6$ µJ energy/pulse, has been used as seed. iii) A pump-probe setup, based on a Ti:Sapphire cavity-dumped oscillator (Coherent Mira 900), to explore different repetition rates, from 250 kHz to 1 MHz. The results reported do not depend on the repetition rate, thus demonstrating that average heating effects are negligible. Measurements with setups i) and ii) were performed at the T-ReX facility at FERMI, Elettra (Trieste). In the setup i), the reflected probe beam is spectrally dispersed through an equilateral SF11 prism and imaged on a Hamamatsu InGaAs linear photodiode array (PDA), capturing the 0.8–2.5 eV spectral region. In configurations ii) and iii) the reflected probe is collected by a commercial Si or InGaAs photodiode. For single-color measurements, lock-in acquisition is used, together with fast ($\sim 60$ kHz) modulation of the pump beam. This ensures a better signal-to-noise ratio with respect to spectroscopic measurements. The sample is mounted on the cold finger of a cryostat. The temperature of the sample is stabilized within $\pm 0.5$ K.

In all experiments, the pump photon energy is 1.55 eV with a fluence ranging from $200 \mu$J·cm$^{-2}$ up to $2.6$ mJ·cm$^{-2}$. The spot sizes of the pump and probe beams on the sample are $\sim 160$ µm and $\sim 100$ µm in diameter, respectively. The data reported in Figs. [2] and [3] have been taken with a pump incident fluence of $300 \mu$J·cm$^{-2}$. Considering the lattice specific heat at 160 K, $C_p \sim 3$ J cm$^{-3}$K$^{-1}$ [19], and estimating the energy absorbed within the 40 nm film via a multi-reflection model (absorption $A=0.18$), we obtain an absorbed energy density $E_{abs}=13$ J/cm$^3$ that corresponds to a temperature increase $\delta T= E_{abs}/C_p \sim 4$ K. The same absorbed energy density corresponds to an absorbed photon density $n_{ph}=5.4 \cdot 10^{19}$ cm$^{-3}$, which can be converted into $1.6 \cdot 10^{-3}$ photons/V atom and an induced orbital polarization $\delta p=3.2 \cdot 10^{-3}$.

D. PEEM image analysis

The analysis of the XMLD-PEEM images have been performed by means of the image processing package Fiji [50][51]. Whithin this package, the Weka Trainable Segmentation [52] machine learning algorithms have been applied to the images to produce pixel-based segmentations, and consequently, to identify different XMLD contrast regions. Once the metallic areas inside a single domain have been highlighted, we calculate the percentage of metallicity by counting the number of recognized pixel with respect to the total pixels number in a fixed domain. A weighted average is evaluated with respect to the size of several domains to give more accurate results.

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Supplementary Information

I. SAMPLE QUALITY

To address the structural quality of the 40 nm V$_2$O$_3$ film epitaxially grown on (0001)-Al$_2$O$_3$, X-ray diffraction measurements were performed on a Panalytical X’pert Pro diffractometer using monochromatic K$_\alpha_1$ radiation of a Cu anode. A film thickness of 40 nm and a surface roughness below 1 nm were determined from X-ray Reflectivity (XRR) measurements and the simulation of the data with the X’pert Reflectivity software. The XRR measurement and simulated curve are shown in Fig. S1(a).

The $\theta$-$2\theta$ scan in Fig. S1(b) shows the symmetric peak (0006) of the film and substrate, thus confirming the single crystalline nature of the film, with the $c$-axis oriented perpendicular to the surface. The finite-size oscillations (Pendellösung fringes) around the film peak maximum confirm the high quality epitaxial deposition with a smooth surface and interface.

Finally, to extract the lattice parameters ($a$ and $c$) of the film, Reciprocal space maps (RSM) around the asymmetric peak (1 0 -1 10) of the film and the substrate were performed. The resulting RSM transformed to in-plane ($a$-axis) and out-of-plane ($c$-axis) lattice spacings is shown in Fig. S1(c). The lattice parameters extracted from the peak maximum are $a$=4.977 Å and $c$=13.962 Å and correspond well with the bulk V$_2$O$_3$ values of $a$= 4.954 Å and $c$=14.008 Å. The small tensile (compressive) in-plane (out-of-plane) strain of 0.46% (-0.33%) is thermal in nature [48].

FIG. S1. (a) XRR measurement and simulated curve to extract the film thickness and surface roughness. (b) XRD $\theta$-2$\theta$ scans around the (0006) reflections of the 40 nm V$_2$O$_3$ film and Al$_2$O$_3$ substrate. (b) RSM transformed to in-plane and out-of-plane lattice spacings of the film and the substrate.

II. DOMAIN ORIENTATION AND PINNING

The XMLD contrast of the insulating domains depends on the relative angle between the direction of the linear horizontal light polarization and the antiferromagnetic axis within each domain. In Fig. S2(a) we report different PEEM images of the same sample area acquired by rotating the V$_2$O$_3$ film around the $c$-axis, i.e. at different angles.
(φ) between the in-plane light polarization and the b-axis. While the morphology of the domains does not change, the XMLD signal (I_{XMLD}) scales as:

\[ I_{XMLD} = A + B \cdot \sin^2(\phi) \]

as expected for magnetic linear dichroism signals [36]. In Fig. S2 b), we report the angle-dependence of \( I_{XMLD} \) of the domain highlighted by the yellow line.

Fig. S3 shows that, after an entire heating-up and cooling-down cycle, the insulating striped domains emerge in the same spatial position and maintain the same shape.

FIG. S2. (a) XMLD-PEEM images at different angles between the in-plane electric field polarization of the X-rays and the crystallographic b-axis of the V2O3 film. The yellow line highlights the domain which has been analyzed in panel (b). (b) \( I_{XMLD} \) as a function of the angle φ between the b-axis and the X-ray polarization direction (double-headed bold red arrow). The insets shows a sketch of the experimental configuration.

FIG. S3. XMLD-PEEM images as a function of the temperature. The insulating striped domains reform in the same position after a heating and cooling cycle. Note the different XMLD contrast between the top and bottom images, which have been taken at different angles between the b-axis and the in-plane light polarization.