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Article

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Extraordinary Hall Effect in Freestanding Oxide Heterostructures

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Abstract:

Recently, Topological Hall Effect (THE) has been unravelled in various perovskite iridate, ruthenate and manganite interfaces, suggesting the presence of magnetic Skyrmion-like bubbles. Among the materials and sample structures investigated so far, the physical properties were not free from influences of substrates, which are templates for crystalline epitaxial growth. Additional contributions of Dzyaloshinskii-Moriya Interaction or Berry curvatures by the substrates may originate from several structural factors such as in-plane strain, octahedral rotation or terminations. In this report, we employ the water-soluble $\text{Sr}_3\text{Al}_2\text{O}_6$ sacrificial buffer layer to prepare freestanding $\text{SrRuO}_3/\text{SrIrO}_3$ heterostructures with high-quality interfaces, and THE-like humps are observed up to 90 K. Analysing the Hall signal with the rotation of magnetic field direction suggests there are magnetically decoupled regions. This scheme also paves the way for several magnetic texture imaging techniques in transmission mode for future studies.

Among the myriad perovskite oxides available across the Periodic Table, the SrRuO_3 (SRO)/ SrIrO_3 (SIO) interface stands out as the robust candidate for exploring the physics of Topological Hall Effect (THE)^{1, 2, 3, 4, 5}. This is true considering their several valuable properties: (i) both the Ru^{4+} ($4d\ t_{2g}^4\ e_g^0$, $L=1$, $S=1$) and Ir^{4+} ($5d\ t_{2g}^5\ e_g^0$, $J_{\text{eff}}=1/2$) are heavy metal cations in perovskite environment contributing strong spin-orbit coupling (SOC). (ii) The bulk SRO has a minority-spin double-exchange mechanism⁶ that creates a metallic ferromagnetic ground state with a net moment of $\sim 1.6\ \mu_B/\text{Ru}$ and strong perpendicular magnetic anisotropy (PMA)⁷; yet the bulk SIO is a paramagnetic semimetal due to its low Mott-Hubbard interaction (U) and Hund's coupling (J_H)^{8, 9}. (iii) SRO and SIO perovskites are blessed with optimal Goldschmidt tolerance factor of ~ 0.9 that facilitates the growth of high quality crystalline ultrathin films and sharp interfaces, easily accessible with a physical vapour deposition technique. Owing to the topologically non-trivial t_{2g} band structure of SRO in k -space¹⁰, factor (ii) produces a large Anomalous Hall Effect (AHE) below its Curie temperature (T_c). Combining factor (i) and (iii) may result in strong interfacial Dzyaloshinskii-Moriya Interaction (DMI)^{11, 12} and may host Neel-type magnetic Skyrmions^{13, 14, 15}, which are distinct to the Bloch-type ones arisen from bulk inversion asymmetry¹⁶.

Thus far, the origin of the hysteretic antisymmetric humps in Hall Effect is subjected to debates. On the one hand, Bruno first derived the theory of THE from a real-space Berry curvature perspective. At a Skyrmion phase, by assuming a strong Hund's coupling between the free electrons' spin and local moments, a Berry phase is gained during the hopping process of free electrons across non-collinear spin textures^{17, 18}. Nakazawa further improvised the theory by accounting for Mott-Hubbard interaction and weak Hund's

coupling¹⁹. Note that this can be generalized to any non-collinear moments^{20, 21} with chirality $\chi_{ijk} = \hat{\mathbf{m}}_i \cdot (\hat{\mathbf{m}}_j \times \hat{\mathbf{m}}_k) \neq 0$, not limited to its integral form of topological charge, $Q = \frac{1}{4\pi} \int \hat{\mathbf{m}} \cdot \left(\frac{d\hat{\mathbf{m}}}{dx} \times \frac{d\hat{\mathbf{m}}}{dy} \right) dx dy = \pm \text{integer}$ (strict definition for real Skyrmions^{13, 22}). THE signal would occur whenever the non-collinear region contributes an “emergent” magnetic field $\vec{\mathbf{B}}_e = \frac{Q\phi_o}{A} \hat{\mathbf{z}}$ in the z-direction, where $\phi_o = \frac{\hbar}{2e}$ is the flux quantum and A is the area. In this work, this (former) is coined as the “AHE+hump” interpretation for any typical extraordinary Hall Effect loop. The apparent AHE can be fitted by one hysteretic Langevin function and saturates at large field, which represents the k-space Berry curvature concept with Chern number¹⁰, $Z_n = \frac{i}{2\pi} \int_{BZ} \nabla_{\mathbf{k}} \times \langle u_{n,\mathbf{k}} | \nabla_{\mathbf{k}} | u_{n,\mathbf{k}} \rangle d^2\mathbf{k}$. Whereas the antisymmetric humps are assumed as the THE signal arisen from non-collinear magnetic textures that vanish at large field. On the other hand, such features are also argued to be an incomplete cancellation of two decoupled Langevin functions:

$$L_{1,2} = A_{1,2} \left\{ \coth \left[\frac{g\mu_o\mu_B J_{1,2}}{k_B T} (H \pm H_{C1,2}) \right] - \frac{k_B T}{g\mu_o\mu_B J_{1,2} (H \pm H_{C1,2})} \right\} \quad (1)$$

where $\text{sgn}(A_1) = -\text{sgn}(A_2)$, $H_{C1} \neq H_{C2}$ and $J_1 \neq J_2$. The latter is coined as the “Bi-Langevin” interpretation here, and it has been perceived to *not* containing THE signal but merely a mathematical artefact from two AHE signals^{23, 24}, either intrinsic or extrinsic. An example is illustrated in supplementary Fig S1a-b: in a naïve magnetic tunnel junction (MTJ) with fully-decoupled top and bottom ferromagnetic layers, a Hall peak may appear at the high-resistance (antiparallel) state, but does not contain any non-collinear/non-coplanar magnetic texture. Recent work on V:(Bi, Sb)₂Te₃ favoured this latter interpretation using a concept of bulk and surface contributions, but did not rule out Skyrmions existence²⁵.

In this report, we present Hall Effect results from freestanding SIO/SRO heterostructures in (001) orientation, prepared via the robust water-soluble $\text{Sr}_3\text{Al}_2\text{O}_6$ (SAO) as a lattice-matched buffer layer for epitaxial lift-off^{26, 27}. We mitigated possible chemical reactions by minimizing the immersion (float) time of flakes in water. Distinct to the epitaxial cases, the hump signals are suppressed in the freestanding bilayer structures yet survive in the trilayer structure, and the negative AHE component prevails. We also analyse the Hall data with the two intensely debated interpretations mentioned above. Finally, the Hall components are observed to have different sensitivity towards the magnetic field direction, suggesting the existence of two magnetically decoupled entities.

First, we compared the properties among three samples structures (Fig. 1a, 3a, 3c), with the details of sample fabrication are clarified in *Methods*. In Figure 1a, the AHE and hump signals are present in both structures A and B, yet their low temperature (ground state) AHE have opposite signs. The low-temperature negative AHE in structure A is consistent with the reported behaviours for SIO/SRO bilayers and SRO single layer^{1, 4}. This can likely be attributed to the minority spin double-exchange mechanism in bulk SRO with threefold-degeneracy (Fig. 1c, left). Since the mobile electron's spin is opposite to the local magnetization, we may expect a negative spin polarization $P_S = \frac{DOS_{\uparrow}(E_F) - DOS_{\downarrow}(E_F)}{DOS_{\uparrow}(E_F) + DOS_{\downarrow}(E_F)}$, which has been reported to be -9.5% at 0.31 K²⁸. However, the AHE sign-reversal at a higher temperature (near T_C) can be understood as thermally-driven E_F shifting across band structure anti-crossings^{29, 30}, which are gaps opened by SOC acting as sources/sinks of Berry curvature^{31, 32}. On the other hand, the ultrathin SRO layer grown on SIO in structure B has more severe carrier localization (Mott-insulating) and d_{xy} orbital is theorized to have

reduced intra-site Coulomb repulsion compared to $d_{xz,yz}$ orbitals, lifting their degeneracy³³,³⁴. The Ru^{4+} electronic configuration is hence fixed at “ferro-orbital” $d_{xz,yz}^{\uparrow}, d_{xz,yz}^{\uparrow}, d_{xy}^{\uparrow\downarrow}$, promoting a $\text{Ru}^{4+}\text{-O}^{2-}\text{-Ru}^{4+}$ antiferromagnetic superexchange (Fig. 1c, middle). While majority mobile electrons flow through the SIO layer in both structures A and B, an electron transfer³⁵ from Ir^{4+} to Ru^{4+} would also occur, contributing an interface ferromagnetic $\text{Ir}^{(4+\delta)+}\text{-O}^{2-}\text{-Ru}^{(4-\delta)+}$ superexchange (Fig. 1c, right) and $P_S > 0$. An *ab-initio* derived tight-binding calculation indeed reported $\text{Ru}^{4+}/\text{Ir}^{4+}$ perovskite interface to have a positive-sign intrinsic AHE contribution³⁶. Note that such $P_S > 0$ state may also dominate in structure A if the bottom SRO thickness is further reduced. More evidence of this concept is shown in supplementary Fig. S1c-d. In structure C, we see a near-complete cancelling of AHE but a strong hump signal up to 100 K, which can be understood as a summation of Hall resistivities of structure A and B. This group of epitaxial structures A, B and C form the basis of our study, and their corresponding freestanding counterparts A_f , B_f and C_f will be discussed in Fig. 3.

Next, the quality of the freestanding structure is carefully optimized. Preliminary experiments indicated prolonged exposure to water might deteriorate the sample quality, probably due to oxygen vacancy migration. Hence, a good strategy is to minimize the float time of samples in water (5-10 minutes), before scooping out onto respective supports. This requires the SAO sacrificial layer to be thick enough (100 nm) for fast water dissolution, yet maintaining layer-by-layer growth with a surface roughness of ~ 0.16 nm. We speculate that the ultrathin SRO/SIO structures A-C be too fragile for convenient scooping during the water dissolution process. Thus a 40-nm-thick STO buffer is inserted above SAO as a mechanical support. Its thickness is optimal to produce elongated flakes

convenient for Hall bar fabrication, above which the flakes would roll along their short axes due to imbalanced surface tension (Fig. 2a), similar to reference³⁷. The rolls would inhibit successful photolithography and Cu electrode lift-off since their Van der Waals adhesion with the SiO₂//Si substrate is weak, hence undesirable. Finally, after transferring onto the SiO₂//Si substrates (Fig. 2b), the flakes maintained a smooth topography of ~ 0.23 nm roughness.

To obtain a sufficiently strong signal in magnetometry measurement, we exfoliated a large (mm-scale) flake of structure C_f by attaching the sample surface to a Kapton tape before immersing into the water; nevertheless, the large flake contains countless cracks due to strain relaxation (Fig. 2c). A clear $T_C \sim 100$ K can be inferred from the moment versus temperature (M - T) curves (Fig. 2d, left panel), where the bifurcation between field-cooled (FC) and zero-field-cooled (ZFC) M - T curves is similar to SRO films having a competition between ferromagnetic and antiferromagnetic interaction between Ru⁴⁺. In the moment versus field (M - H) loops (Fig. 2d, right panel), clear double-hysteresis loops can be seen at a low temperature of 20 K, with the wider coercive field $\mu_0 H_C \sim 1$ T, which is similar to the H_C of Hall Effect to be discussed in Fig. 3g. Thus the wider H_C can be attributed to the well-known Ru⁴⁺/Ru⁴⁺ ferromagnetic exchange, while the narrower $\mu_0 H_C \sim 28$ mT could originate from Ru⁴⁺/Ir⁴⁺ superexchange at the interface, weakly-magnetized Ir⁴⁺ moment by proximity effect with SRO, or oxygen vacancy in the 40-nm STO buffer. The bigger H_C quickly diminishes with temperature and becomes indistinguishable at ~ 60 K, but the thinner hysteresis loop is almost temperature-insensitive and persists up to 300 K. Clear out-of-plane magnetic anisotropy can be seen in both M - T and M - H curves. To justify our choice of the sacrificial layer, we replaced the SAO sacrificial layer with La_{0.67}Sr_{0.33}MnO₃

(LSMO), which is also highly compatible to perovskites and can be etched by a “KI+HCl+H₂O” reducing agent^{38,39} (supplementary Fig. S2). Surprisingly, large continuous flakes can be produced with few cracks, yet the magnetic property is completely diminished, likely due to chemical reaction with the acid.

Next, Cu electrodes are patterned onto freestanding flakes A_f , B_f and C_f by standard photolithography and lift-off, as shown in Fig. 3a-b. Apparently, from Fig. 3c, structure A_f (B_f) is similar to A (B) in terms of their respective negative (positive) AHE signs. Yet, the humps are suppressed at all temperature, and structure B_f has slightly larger H_c than that of structure B, A and A_f at 10 K, indicating more defects-pinning of the top SRO. Surprisingly, the humps can be observed in structure C_f up to 80 K, together with the dominant negative-sign AHE component. It is reasonable to infer that the Hall signal of structure C_f is, again, an addition of the Hall signals from A_f and B_f , thus in favour of the “Bi-Langevin” interpretation. An intermediate state of structure C_f before water-immersion is shown in supplementary Fig S3a-b. We can further infer that removing the STO substrate results in the elimination of a positive-sign intrinsic AHE, which possibly originates either from the Ru/Ti interface in agreement to the tight-binding calculation of reference ³⁶. On the other hand, since the STO substrate is cubic, the octahedral rotation of a fraction of SRO adjacent to the substrate will be restricted into tetragonal phase with a weaker moment, contributing positive-sign AHE^{4, 40}; which, in turns, to be eliminated upon removal of the substrate. In short, the competing contributions of intrinsic AHE could be qualitatively summarized as follow, referring to the reasonable $\sim 1.0 \mu_B/\text{Ru}$ regime for ultrathin SRO:

Contributions	AHE Sign	Presence in structures
Ru/Ti	+	A, C
Ru/Ru	-	A, B, C, A _f , C _f
Ir/Ru	+	B, C, B _f , C _f

Note that in Fig. 3d, the humps persist only up to 40 K for the case of 180-minute water-immersion before scoop-out and can be understood as a weakening of the bottom SRO ferromagnetism. This justifies our afore-mentioned strategy that reducing the water immersion time significantly improves the data consistency, by mitigating the structural/chemical damage to the bottom SRO. Besides, the Ordinary Hall Effect ($\rho_{xy}^{\text{OHE}} = R_o B \approx \frac{1}{ne}$) gradient becomes smaller for all freestanding flakes compared to the epitaxial ones, probably due to oxygen vacancy in the 40-nm STO buffer contributing higher carrier density (supplementary Fig. S3c-d).

The “Bi-Langevin” interpretation is perhaps incomplete to decipher the mystery of the hump signals – doubts still arise in the reason of decoupling between $L_{1,2}$ at the hump’s emergence, and their sensitivity towards the magnetic field direction. Therefore, it would be insightful to study the Hall Effect evolution by rotating the external magnetic field direction from out-of-plane (H_z , $\theta=0^\circ$) to in-plane (H_y , $\theta=90^\circ$) (schematic in Fig. 4b, inset). As seen in Fig. 4a-b, we analyse the angle-dependence of the extraordinary Hall components extracted from supplementary Fig. S4 with both the “AHE+hump” and “Bi-Langevin” interpretations. Using the former, the humps vanish at $\theta\sim 50^\circ$, accompanied by a pronounced increase of the apparent net AHE before diminishing again at $\theta>70^\circ$. Conversely, with the latter by fitting to Eqn. (1), we see a fast suppression of L_2 also upon reaching $\theta\sim 50^\circ$, yet the larger L_1 remain almost unchanged until $\theta>70^\circ$. On the other hand,

$H_{C,AHE}$ (of the former interpretation) and H_{C1} (latter) gradually increases by a trivial $1/\cos(\theta)$ trend, showing an apparent out-of-plane anisotropy. Yet, the humps' peak field ($H_{P,hump}$) (former) and the L_2 coercive field H_{C2} (latter) remains unchanged with increasing θ , indicating the absence of anisotropy.

We recall from Ginsburg-Landau framework⁴¹, Bloch-type magnetic Skyrmions lattice (SkL) is formed by intersecting a trio of helicoids (3q-state) with wave-vectors in xy-plane mutually subtended at 120° . Its reliance on z-direction magnetic field is strict – an xy-plane field would disturb the trio's balance, transforming the SkL phase back into helical stripe domains^{42, 43, 44}. Using micromagnetic simulations, we illustrate this concept in Neel-type Skyrmions via the MUMAX3 platform⁴⁵, as shown in Fig. 4c. The parameters used in the simulation are specifically chosen for ultrathin SRO at 80 K, and are justified in the supplementary text and Fig. S5. The agreement in trend and critical $\theta=70^\circ$ between the simulated topological charge density $TCD = \hat{\mathbf{m}} \cdot \left(\frac{\partial \hat{\mathbf{m}}}{\partial x} \times \frac{\partial \hat{\mathbf{m}}}{\partial y} \right)$ with the measured hump features suggests that the humps can indeed be interpreted as THE signals for the emergence of topologically non-trivial magnetic textures such as Skyrmions carrying a net Q . Such textures are expected to show different behaviour compared to collinear domains. Conversely, the example of two fully-decoupled collinear-magnetized bodies as illustrated in supplementary Fig. S1a is expected to have both $|A_{1,2}|$ vanishing at matching θ and both $H_{C1,2}$ following the same $1/\cos(\theta)$ trend upon deconvolution of its Hall signal. This way, the apparent humps should gradually shift to larger θ but is not expected to vanish at smaller θ , as illustrated in supplementary Fig. S6b. The rotation of the magnetic field towards in-plane may prove to be a robust tool for distinguishing such decoupled $L_{1,2}$ in future work.

In real-space magnetic texture imaging, The Lorentz Transmission Electron Microscopy (LTEM) relying on the intensity contrast⁴⁶ $I \propto (\nabla \times \hat{\mathbf{M}}) \cdot \hat{\mathbf{k}}$ remains the hitherto most convincing technique in verifying the existence of Bloch-type Skyrmions^{16, 47, 48}, where $\hat{\mathbf{k}}$ is the electron propagation vector. However, for Neel-type Skyrmions (typically thin films on substrates), since the mentioned contrast is inaccessible, several scanning probe techniques have been employed instead, e.g. for the monolayer Fe//Ir(111)⁴⁹ and GaV₄S₈⁵⁰. Notably, some cryogenic Magnetic Force Microscopy (MFM) provided evidence of magnetic bubbles corresponding to THE signal emergence in SRO/SIO epitaxial bilayers^{1, 51, 52, 53}. However, we believe there is room for improvement. To this end, our freestanding oxide flakes transferred onto nm-thick Si₃N₄ membranes may pave the way for future complementary imaging studies by LTEM with sample-tilt⁵⁴, or Scanning Transmission X-ray Microscopy (STXM)⁵⁵.

In conclusion, using SAO buffer, we fabricated freestanding SRO/SIO flakes and demonstrated preservation of high-quality interfaces that support extraordinary Hall Effect, by minimizing the immersion time in the water. Rotating the magnetic field direction revealed new insights towards Hall Effect analyses. Removing the substrates may prove to be a viable strategy in facilitating more fundamental research in various branches of crystalline condensed matter Physics.

Methods:

Sample fabrications

STO(001) substrates were etched with buffered HF solution and annealed at 950 °C to achieve single TiO₂-termination. All films involved were grown in pulsed laser deposition chambers. To define the Hall bars for the structure A, B, C and C_f-intermediate, the substrates were first patterned by photolithography followed by amorphous AlN deposition at room temperature and base vacuum. This eliminates the possibility of introducing oxygen vacancies into the STO substrates by standard Argon ion milling that might create unwanted complications. Hence, we could exclude analyses of impurity/disorder-induced extrinsic AHE mechanisms such as skew scattering or side-jump⁵⁶. All single-crystalline SRO and SIO films were grown at 650°C, 0.14 mbar of oxygen, and 2 J/cm² of laser energy density. Such parameter should result in majority monoclinic-phase^{4, 40} of SRO (Glazer notation a⁻b⁺c⁻), as distinct to the tetragonal-phase (a⁰a⁰c⁻). Hence, our single-layer ultrathin SRO does not produce the debated hump features in Hall Effect within measurable range. For structures A_f, B_f and C_f, 100 nm-thick SAO films were grown at 800 °C and base vacuum (5x10⁻⁶ mbar); followed by 40 nm-thick STO buffer at 800 °C and 5x10⁻⁴ mbar to ensure layer-by-layer growth.

Characterizations

All Ordinary Hall Effect (OHE) background, which is linear with the magnetic field, has been removed from the Hall Effect data presented in the main text figures, and some of them are presented in supplementary Fig. S3c-d. Rotation of the magnetic field is done using a Quantum Design PPMS sample rotator setup. The magnetic moment is measured by a Quantum Design MPMS3 SQUID-VSM. Since the contributions of oxygen vacancy to

electrical transport and magnetic moment are difficult to quantify, the contribution of the 40 nm-thick STO buffer is excluded from the conversion of raw data of resistance into resistivity and magnetic moment into emu/cc.

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Author contributions:

A.A. and Z.S.L designed the research ideas. Z.S.L. and Z.Z. performed the experiments. Z.S.L., Z.Z. and A.A. drafted the manuscript. The other authors contributed valuable advice during discussions.

Competing Interest:

The authors declare no competing interest.

Data availability:

The data that support the findings of this study are available from the corresponding author on request.

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Figures:

Figure 1: Hall Effect data of the epitaxial basis. (a) Schematics of structures A, B and C and their surface topography attached. **(b)** The corresponding Hall Effect data from the three epitaxial structures, with dotted lines indicating the hysteresis direction. **(c)** Illustrations of (oxygen-mediated) threefold-degenerate ferromagnetic minority-spin double-exchange (left panel), and “ferro-orbital” antiferromagnetic superexchange (middle panel) between neighbouring Ru^{4+} ions in perovskite environment. Right panel: Proposed $\text{Ru}^{4+}/\text{Ir}^{4+}$ interfacial ferromagnetic superexchange.

Figure 2: Flakes' morphology and magnetic properties. Optical microscopy images of curled-up roll **(a)** and flat flake **(b)**, adhered on SiO₂//Si support. **(c)** Optical microscopy image of an aggregate of cracked flakes exfoliated by a polyimide (Kapton) tape, to be used for generating the M - T (left) and M - H curves (right) shown in **(d)**. The M - H curves of different temperatures are shifted vertically for clarity.

Figure 3: Hall Effect data of the freestanding flakes. (a) Schematics of structure A_f, B_f and C_f with topography attached. (b) Optical microscopy image of a flake patterned with Cu electrodes. Hall Effect data for (c) structure A_f (left), B_f (middle) and C_f (right) after minimal float time, and (d) for structure C_f after long float time. The data in (c) and (d) are shifted vertically for clarity.

Figure 4: Hall Effect of the structure C_7 at 80 K with magnetic field rotation towards in-plane. θ -dependence of the absolute magnitude of **(a)** the extraordinary Hall components ($|AHE|$, $|hump|$, $|A_1|$, $|A_2|$), and **(b)** their coercive fields ($H_{C,AHE}$, H_{C1} , H_{C2}) and humps' peak field ($H_{P,hump}$) after deconvolutions according to the two interpretations. $A_{1,2}$ are the coefficients of $L_{1,2}$ (Eqn. 1). The coloured dashed lines in (a) are extrapolations. The magenta dotted line in (b) is a guide-to-the-eye for $\propto 1/\cos(\theta)$ trend. **(c)** Simulated θ -dependent TCD, corresponding to domain textures shown in the surrounding snapshots.

Figures

Figure 1

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Figure 4

Hall Effect of the structure Cf at 80 K with magnetic field rotation towards in-plane. θ -dependence of the absolute magnitude of (a) the extraordinary Hall components ($|AHE|$, $|hump|$, $|A_1|$, $|A_2|$), and (b) their coercive fields (H_C , AHE, H_{C1} , H_{C2}) and humps' peak field (H_P , hump) after deconvolutions according to the two interpretations. $A_{1,2}$ are the coefficients of $L_{1,2}$ (Eqn. 1). The coloured dashed lines in (a) are extrapolations. The magenta dotted line in (b) is a guide-to-the-eye for $1/\cos(\theta)$ trend. (c) Simulated θ -dependent TCD, corresponding to domain textures shown in the surrounding snapshots.

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