

Three-Dimensional Visualization of Conductive Filaments in TiO_2 via Conductive Atomic Force Microscopy Tomography

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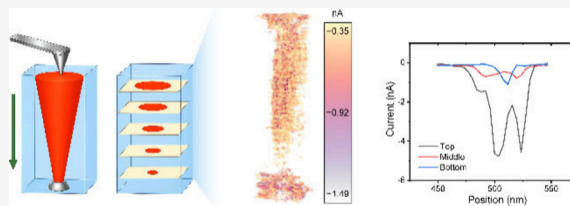
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Supporting Information

ABSTRACT: Direct three-dimensional visualization of conductive filaments (CFs) in valence change memory is essential for resolving whether their spatial evolution is governed by stochastic defect generation or by electrode geometry. Here, we apply conductive atomic force microscopy tomography to a TiO_2 (10 nm)/Pt structure, extending this technique to TiO_2 -based CFs previously inaccessible to tomographic characterization. The tomogram reveals a reverse-cone morphology whose terminus preferentially connects to a Pt bottom electrode (BE) protrusion, evidencing dual field enhancement at both interfaces. Current gradient analysis reveals a nonmonotonic conductivity profile, with values of 0.1355, 0.0682, and 0.1035 nA/nm at the top, middle, and bottom, consistent with lateral electrostatic confinement by the BE protrusion. Competing exponential field dependence of vacancy generation and linear field dependence of drift accounts for the near-tip constriction. These results establish the BE morphology as an active confinement mechanism and identify nanoscale BE roughness as a key parameter influencing switching variability in resistive random access memory.

KEYWORDS: Conductive Filament, C-AFM Tomography, Resistive Random Access Memory (RRAM), Resistive Switching, Dual Field Enhancement



Resistive random-access memory (RRAM) has emerged as a leading candidate for next-generation nonvolatile memory owing to its simple device structure, fast switching speed, and high integration density.^{1–4} Among RRAM variants, valence change memory (VCM) based on transition metal oxides such as TiO_2 operate through the reversible formation and rupture of nanoscale conductive filaments (CFs) composed of oxygen vacancies.⁵ Although CF-based switching has been studied extensively, the widespread commercialization of RRAM remains hindered by variability in SET/RESET parameters and limited endurance.⁶ These reliability issues stem from an incomplete understanding of whether CF morphology and spatial evolution are genuinely stochastic or are governed by electrode interface geometry.⁷ Resolving this question demands direct, three-dimensional (3D) characterization of individual CFs under well-controlled conditions.

To access the 3D structure of CFs, conductive atomic force microscopy (C-AFM) tomography—also termed scalpel scanning probe microscopy⁸—has been established as a powerful tool. Seminal studies employing this technique revealed forward-cone morphologies and quantized conductance behavior in metallic filaments formed in conductive-bridging RAM (CBRAM), or electrochemical metallization memory (ECM), systems.⁹ However, applying C-AFM tomography to VCM systems is inherently more challenging: oxygen-vacancy-based CFs exhibit significantly lower conductivity than their metallic ECM counterparts, yielding a weaker signal-to-noise ratio that complicates 3D imaging.

While prior work has explored VCM filament structures in materials such as SiO_x , the intrinsic 3D geometry of a single VCM filament remains poorly understood relative to ECM counterparts.^{10,11} A critical gap therefore persists: direct 3D visualization of how the CF growth path interacts with the bottom electrode (BE) interfacial morphology—a factor widely recognized to influence switching behavior and reliability—is absent.^{12,13} Moreover, existing tomography studies have focused predominantly on exterior morphological reconstruction, leaving the internal conductivity distribution and its correlation with local electric field dynamics largely unexplored.

Here, we apply C-AFM tomography using a conductive diamond tip to a TiO_2 (10 nm)/Pt structure. By forming the filament with an AFM tip under a highly localized electric field, we overcome the challenge of low conductivity contrast by generating a more clearly distinguishable filament and precisely defining its position. This approach enables the first full 3D reconstruction of the morphology and internal conductivity distribution of a single TiO_2 -based CF.

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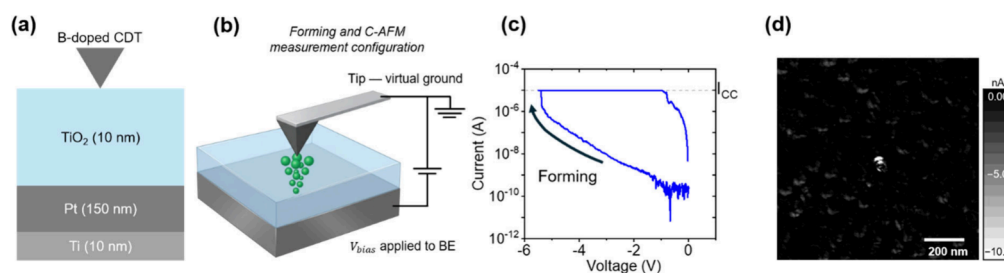


Figure 1. Sample structure and electrical characterization of the TiO₂/Pt system. (a) Schematic illustration of the device structure consisting of a 10 nm amorphous TiO₂ switching layer on a 150 nm Pt BE. (b) Schematic of the C-AFM measurement setup, showing the boron-doped diamond tip in contact with the TiO₂ surface and the circuit configuration for voltage application and current compliance control. (c) *I*–*V* curve recorded during the forming process, showing switching to the LRS at a compliance current of 10 μA. (d) C-AFM current map of a 1 × 1 μm² area after forming, revealing a localized high-conductivity region with a diameter of less than 50 nm, confirming the formation of a single CF.

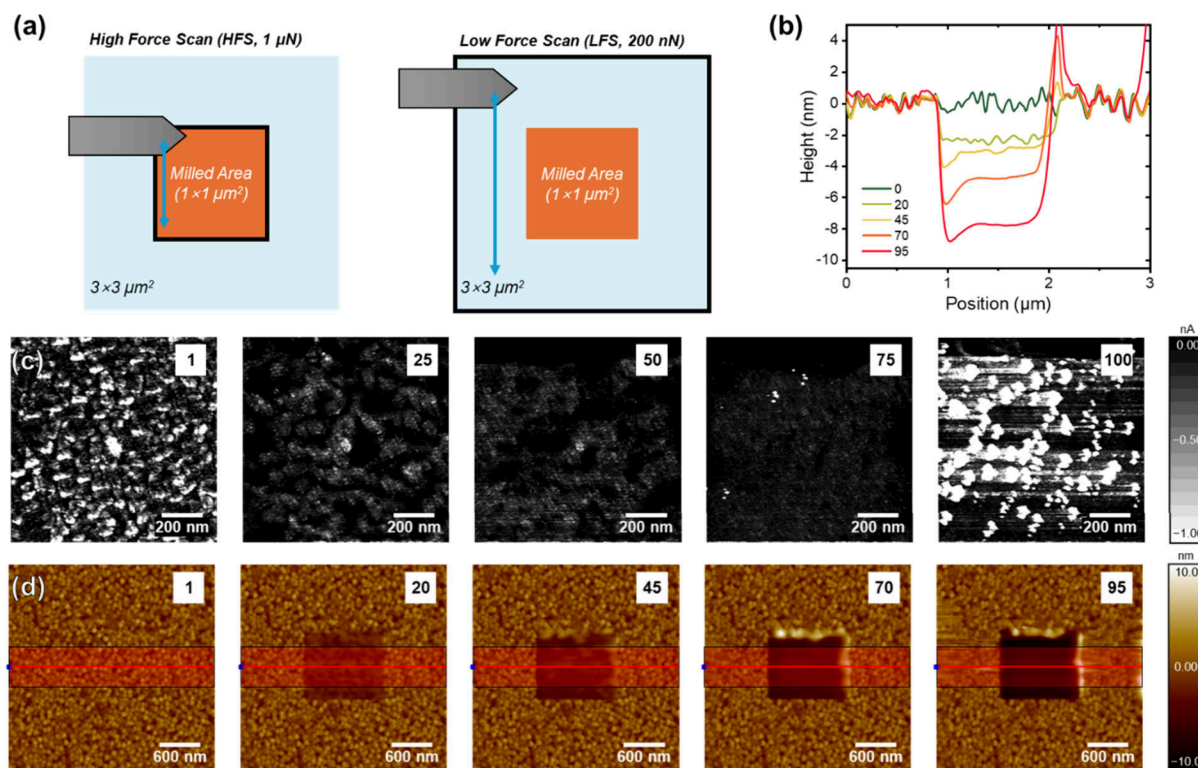


Figure 2. C-AFM tomography procedure and milling characterization. (a) Schematic of the alternating LFS/HFS cycling protocol used for controlled layer-by-layer milling. (b) Height line profiles extracted from the red-boxed regions in part d, showing a steady monotonic decrease at an average milling rate of 0.1 nm per scan. (c) Representative C-AFM current maps (sample bias 300 mV) acquired after 1, 25, 50, 75, and 100 HFSs, showing the evolution of the CF signal with progressive milling depth. (d) Corresponding AFM height images obtained during LFSs (sample bias 500 mV) at cycles 1, 20, 45, 70, and 95, confirming a flat and artifact-free milled surface throughout the process.

The reconstructed 3D tomogram reveals a distinct reverse-cone morphology, wider near the tip and narrowing toward the BE. Crucially, the 3D visualization provides direct evidence that the CF terminus connects to a specific nanoscale protrusion on the Pt BE surface, demonstrating that CF spatial evolution is preferentially guided by a dual field-enhancement mechanism: top-down filament growth driven by the intense electric field at the tip–TiO₂ interface, synergistically guided by localized field enhancement at the BE protrusion.^{14,15}

Furthermore, we extend C-AFM tomography beyond morphological reconstruction by quantitatively analyzing the internal conductivity distribution through Gaussian fitting of lateral current profiles at each milling depth. The resulting current gradient map reveals a nonmonotonic distribution:

steep lateral confinement at both the top and bottom interfaces with a comparatively diffuse boundary in the middle region. The reshaping of the current profile at the BE protrusion provides direct experimental evidence that BE interfacial morphology acts as an active spatial confinement mechanism, ultimately governing the structural integrity and precise location of the CF.

The sample consists of a 10 nm amorphous TiO₂ switching layer deposited on a 150 nm Pt BE, as illustrated in Figure 1a,b. A boron-doped conductive diamond tip served as the mobile top electrode (TE) for both *I*–*V* spectroscopy and C-AFM current mapping. The forming process was initiated by sweeping the voltage from 0 to –5.5 V applied to the BE. Switching to the low resistance state (LRS) was confirmed when the current reached the compliance limit of 10 μA at

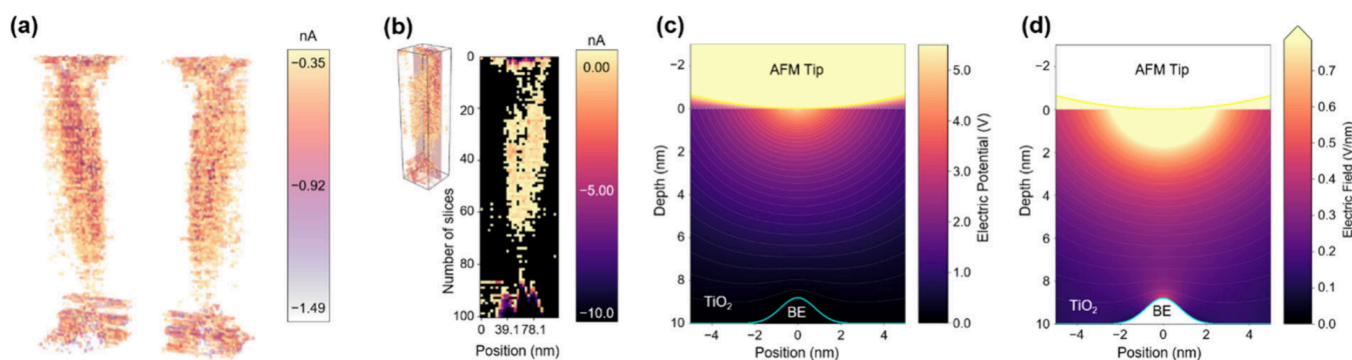


Figure 3. 3D reconstruction and dual field-enhancement mechanism of the CF. (a) Front and rear views of the fully reconstructed 3D tomogram, revealing a reverse-cone morphology wider near the tip and narrowing toward the BE. (b) Cross-sectional view of the tomogram, showing the physical connection between the CF terminus and a specific nanoscale protrusion on the BE surface. (c) FDM-simulated electric potential distribution around a Gaussian-profiled BE surface protrusion. (d) Calculated electric field magnitude, revealing localized field enhancement at the BE protrusion, confirming its role as a secondary field concentrator that guides the CF terminus.

−5.5 V; upon sweeping back toward 0 V, the current remained at the compliance limit until approximately −1 V, as shown in the I – V curve in Figure 1c. To spatially resolve the conductivity change, the $1 \times 1 \mu\text{m}^2$ area surrounding the voltage application site was subsequently scanned by C-AFM. The resulting current map (Figure 1d) reveals a localized high-conductivity region—a bright spot at the center with a diameter of less than 50 nm—confirming the formation of a single CF.

To resolve the vertical structure of the CF, C-AFM tomography was performed using the conductive diamond tip as a nanoscale scalpel while simultaneously mapping local conductivity. The procedure alternated between one low-force scan (LFS) at 200 nN and five high-force scans (HFS) at $1 \mu\text{N}$, as illustrated in Figure 2a. This cycling strategy ensured milling consistency and enabled the milling rate to be monitored every five cycles, providing precise control over material removal depth. Figure 2c shows representative C-AFM current maps acquired after 1, 25, 50, 75, and 100 HFSs, and Figure 2d shows corresponding height images from LFSs at cycles 1, 20, 45, 70, and 95. The milled surface remained flat and free of significant roughness throughout the process. Over 100 HFSs, the entire 10 nm TiO_2 layer was removed at a stable average milling rate of 0.1 nm per scan. Consistent current signals in the central region of the HFS maps confirm complete CF formation during the SET process. The height line profiles in Figure 2b, derived from the red-boxed regions in Figure 2d, show a steady monotonic decrease in height, validating the gradual and predictable oxide removal. This reliable depth control, achieved through periodic LFS monitoring, is a prerequisite for the z -directional resolution required for high-fidelity 3D CF reconstruction.

Figure 3a presents the front and rear views of the fully reconstructed CF tomogram, and Figure 3b shows the corresponding cross-sectional view. The CF exhibits a reverse-cone morphology, wider near the TE (AFM tip) and narrowing toward the BE. Notably, the cross-sectional view reveals that the CF terminus connects directly to a specific nanoscale protrusion on the BE surface. This protrusion-guided termination is a reproducible feature: it is illustrated for representative filaments in Figure S1 and was observed in five CFs formed at independent locations on the thin film, confirming that the preferential connection between the CF

base and a BE protrusion is a reproducible structural characteristic rather than a coincidental observation.

The reverse-cone morphology is consistent with the power-limited forming conditions employed here and can be understood within a unified framework that reconciles prior reports of varying CF geometries. Studies employing high-compliance currents in the mA range reported conventional conical or dual-conical morphologies, attributable to Joule heating-driven lateral expansion.^{16,17} By contrast, the present study used a compliance current of $10 \mu\text{A}$ —more than an order of magnitude lower—effectively suppressing thermal contributions and placing the forming process in a tip-field-dominated, power-limited regime. Under these conditions, the point-source electric field at the tip apex governs top-down filament propagation, naturally producing a morphology widest at the field origin and narrowing toward the BE. The CF diameter observed here (<50 nm) is consistent with the sub-50 nm filament dimensions reported in a comparable Pt/ TiO_2 system by cross-sectional transmission electron microscopy, providing independent cross-validation of the present tomographic reconstruction.⁵

The nanoscale tip geometry creates a highly concentrated electric field at its apex, driving oxygen ion migration to the surface and leaving behind oxygen vacancies within the oxide. We propose that CF growth initiates at the tip– TiO_2 interface, where the field is most intense, and propagates downward toward the BE in a top-down manner consistent with the observed reverse-cone morphology. The physical connection between the CF terminus and a BE protrusion indicates that field concentration is not limited to the tip– TiO_2 interface but also arises at morphological asperities on the BE surface. This valence-change (anionic) process forms an oxygen-deficient, Magnéli-type conductive channel⁵ rather than a metal electrodeposit;¹⁸ under our forming polarity the tip acts as the positive electrode, so oxygen ions are extracted toward the tip and the resulting oxygen-vacancy chain, whose donor electrons render the channel conductive, propagates the filament.^{1,19} Electron injection becomes relevant under the reverse (reset) polarity, where it neutralizes charged vacancies and promotes reoxidation,²⁰ and therefore does not set the as-formed reverse-cone morphology.

To verify this field enhancement, we performed a finite-difference method (FDM) simulation of the electric potential distribution, shown in Figure 3c. The calculated field

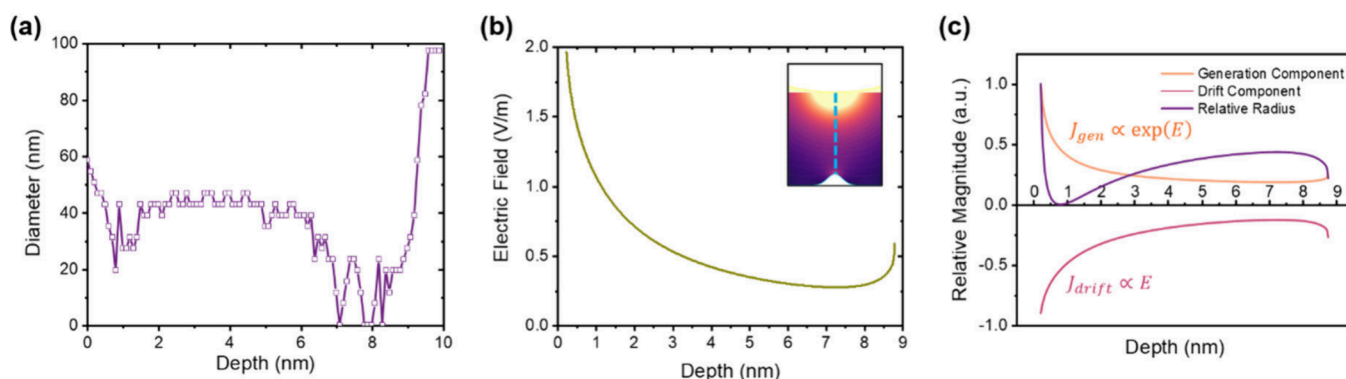


Figure 4. Quantitative diameter profile and necking mechanism of the CF. (a) Depth-wise diameter profile extracted using a current threshold of 150 pA, revealing a nonmonotonic evolution with a sharp constriction at approximately 0.8 nm depth and an abrupt increase near 8.8 nm attributed to tip contact with the 1.2 nm-tall BE protrusion. (b) FDM-simulated z -directional electric field profile along the CF growth axis. (c) Schematic plot of the semiquantitative competition model illustrating the crossover between the exponential field dependence of oxygen vacancy generation flux (J_{gen}) and the linear field dependence of drift flux (J_{drift}) as a function of depth, predicting the existence of a constriction at the crossover depth.

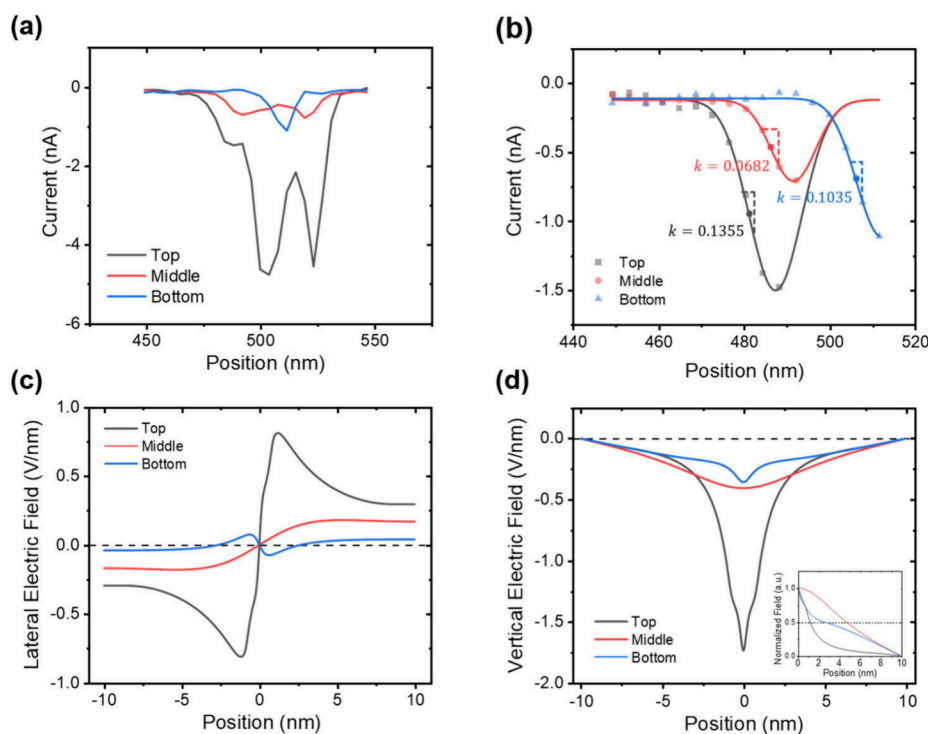


Figure 5. Internal conductivity distribution and electrostatic confinement analysis. (a) Cross-sectional current line profiles extracted from the top (-0.1 nm), middle (-4.2 nm), and bottom (-8.5 nm) regions of the reconstructed tomogram. (b) Current gradient profiles derived by Gaussian fitting of the CF edges at each depth; maximum gradient values of 0.1355, 0.0682, and 0.1035 nA/nm for the top, middle, and bottom regions, respectively, serving as an indicator of internal oxygen vacancy confinement. (c) Lateral electric field component (E_x) at the three depth positions, showing an outward (negative) force in the top and middle regions and a localized inward (positive) force near the BE interface. (d) Vertical electric field component (E_z) at the three depth positions. The inset shows normalized E_z profiles with lateral half-maximum positions of 1.22, 4.74, and 2.56 nm for the top, middle, and bottom regions, respectively, inversely correlated with the current gradient trend.

magnitude in Figure 3d reveals a significant increase near a 1.2 nm-tall protrusion, confirming that concentrated fields at these sites provide a secondary driving force that guides the CF terminus to a specific BE location. Taken together, for this single, tip-formed filament our direct 3D observations indicate that CF spatial evolution—including morphology and growth path—is preferentially guided by the dual field enhancement at both electrode interfaces, a morphological correlation consistent with the enhanced local field.

To elucidate the spatial evolution of CF morphology, we performed quantitative radius analysis as a function of milling

depth. Figure 4a shows the depth-wise diameter profile extracted using a current threshold of 150 pA. The profile reveals a generally decreasing trend down to a depth of approximately 8.8 nm, beyond which the apparent diameter increases abruptly due to tip contact with the 1.2-nm-tall BE protrusion. Notably, it also reveals a distinct nonlinear evolution with a sharp constriction reaching a minimum at approximately 0.8 nm depth. This deviation from a simple inverted-cone model suggests a complex interplay between localized defect chemistry and field-driven transport. The near-tip constriction was reproduced in two additional independ-

ently analyzed filaments (Figure S1c), confirming that necking is a robust morphological feature of the CF population rather than an artifact of a single measurement.

To rationalize this nonmonotonic profile, we performed FDM-based electric field simulations; the z -directional field profile along the CF growth axis is shown in Figure 4b. We propose a semiquantitative competition model in which the CF diameter is governed by the balance between an oxygen vacancy generation flux J_{gen} that drives lateral expansion, and a drift flux J_{drift} that transports vacancies axially along the local field. Because J_{gen} depends exponentially on the local field E owing to field-assisted lowering of the hopping barrier,^{21,22} whereas J_{drift} scales linearly with E ,^{1,23} a critical crossover depth z_c necessarily exists at which the two fluxes balance. As illustrated schematically in Figure 4c, above z_c , where the field is highest near the CF tip, J_{gen} dominates and the filament widens; below z_c , J_{drift} outpaces J_{gen} , leading to net vacancy removal from the filament periphery and a diameter minimum. The FDM-simulated $E(z)$ exhibits its steepest decay within the first ~ 1 nm below the tip apex, consistent with the constriction minimum at ~ 0.8 nm depth. A full description of the model, including parameter selection and sensitivity analysis, is provided in Note S2.

The internal lateral current distribution provides insight into carrier transport dynamics and field confinement. To quantify this, cross-sectional current line profiles were extracted from the top (-0.1 nm), middle (-4.2 nm), and bottom (-8.5 nm) regions of the reconstructed tomogram, as shown in Figure 5a. Figure 5b presents current gradients derived by Gaussian fitting of the CF edges at each depth, with the maximum gradient calculated as $\text{Gradient}_{\text{max}} = A/\sigma\sqrt{e} \approx A/(1.6487\sigma)$. The gradient steepness follows the order top (0.1355 nA/nm) > bottom (0.1035 nA/nm) > middle (0.0682 nA/nm), serving as an indirect indicator of internal oxygen vacancy distribution: densely confined at the interfaces and diffuse in the bulk middle region.

To elucidate the origin of this gradient trend, we calculated the lateral (E_x) and vertical (E_z) electric field components from the spatial derivatives of the FDM potential. The lateral field E_x in Figure 5c governs the directional dynamics of oxygen vacancy migration. Its absolute amplitude follows the order top > middle > bottom. In the top and middle regions, E_x is negative, indicating an outward electrostatic force that promotes lateral expansion of the CF and contributes to the diffuse boundary in the middle region. Near the bottom interface, however, E_x exhibits localized positive regions, generating an inward force that confines diffusing oxygen vacancies toward the filament center.

The vertical field E_z in Figure 5d decreases in absolute amplitude in the order top > middle > bottom. The normalized field profiles in the inset reveal lateral half-maximum positions of 1.22, 4.74, and 2.56 nm at the top, middle, and bottom regions, respectively—a spatial spread that follows the order middle > bottom > top, inversely proportional to the current gradient trend. As the CF approaches the BE, the localized vertical and inward lateral fields generated by the high-aspect-ratio protrusion act as secondary concentrators, actively reconfining diffusing oxygen vacancies and steepening the internal current gradient to match the narrowed exterior morphology. These findings indicate that nanoscale BE interfacial roughness is an active structural factor in the local confinement of the filament, and a relevant parameter for switching reliability in RRAM.

In conclusion, we directly visualized the 3D morphology of a single tip-induced CF in a TiO_2 thin film using C-AFM tomography. The reconstructed tomogram and FDM simulations indicate that, for this single tip-formed filament, CF spatial evolution is preferentially guided by the electrode interface geometry. The CF exhibits a reverse-cone morphology whose origin is unified within a compliance-dependent framework reconciling prior reports across different forming conditions. A distinct nonlinear constriction near the top interface is elucidated by a semiquantitative competition model in which the exponential field dependence of J_{gen} and the linear dependence of J_{drift} necessarily cross at a critical depth under power-limited, low-compliance forming.²⁴ Internal current gradient analysis further reveals that nanoscale BE protrusions act as secondary field concentrators, generating inward lateral electrostatic forces that actively reconfine diffusing oxygen vacancies as the CF approaches the substrate. We emphasize that this preferential, morphologically guided termination concerns where a single filament ends within an individual device and should not be conflated with the cell-to-cell switching variability of a device array. These findings identify nanoscale BE interfacial roughness as an active factor in CF confinement, and highlight BE topography as a parameter worth considering in the design of RRAM devices.

METHODS

TiO_2 thin films were deposited on Pt (150 nm)/Ti (10 nm)/ SiO_2 (300 nm)/Si substrates (Quintess Co. Ltd., Korea) by plasma-enhanced atomic layer deposition (PE-ALD) at 300 °C. Tetrakis-(dimethylamido)titanium (TDMAT) maintained at 40 °C was used as the Ti precursor, with oxygen plasma as the reactant.

All electrical measurements and C-AFM operations were conducted using a commercial C-AFM system (Cypher, Asylum Research, Oxford Instruments) with a current amplifier (ORCA holder) and a solid single-crystal boron-doped diamond conical probe (Adama Innovations, FM-LC; nominal apex radius 20 nm, full cone angle 90°, spring constant ~ 10 N/m) as the mobile TE. All biases were applied to the BE through the sample while the tip was held at the virtual ground of the current amplifier, so that a negative bias denotes a negative potential on the BE relative to the tip. CF formation was induced by applying a negative voltage sweep from 0 to -5.5 V to the BE while the tip was in contact with the amorphous TiO_2 surface. Switching from the high resistance state to the low resistance state was confirmed when the current reached the compliance limit of $I_{\text{cc}} = 10$ μA . Contact forces were calibrated for each probe: the deflection sensitivity (InvOLS) was obtained from a force–distance curve on a rigid sapphire substrate and the cantilever spring constant from the thermal-tune method, and the 200 nN (low-force) and 1 μN (high-force) set points were computed from the calibrated deflection. A solid single-crystal diamond probe was selected because milling-based tomography demands a mechanically durable, wear-resistant scalpel: Pt-coated and solid Pt tips deform or wear under repeated high-force contact scanning—Pt-wire (solid Pt) tips have been observed to bend or wear after only a few consecutive scanning experiments, whereas conducting diamond tips remained essentially unworn²⁵—which would compromise the depth-consistent current mapping the reconstruction depends on.

For C-AFM tomography, the conductive diamond tip was used as a nanoscale scalpel to progressively mill the TiO_2 layer while simultaneously mapping local conductivity. Each HFS (contact force 1 μN , sample bias 300 mV) was alternated with a LFS (200 nN, sample bias 500 mV) that imaged the surface topography; 100 repeated HFSs over the CF formation site yielded 100 sequential 2D current maps as a function of milling depth. All biases were applied through the sample with the tip at virtual ground. Complete removal of the 10 nm TiO_2 layer was confirmed by the appearance of saturated high-current signals corresponding to exposure of the Pt BE.

For 3D reconstruction, a current threshold of -500 pA was applied to the stack of 100 current maps to isolate the CF signal from background noise (measurements were conducted under negative bias, where more negative current corresponds to higher conductivity). Pixels below this threshold were assigned as background; those above were identified as the CF. The processed 2D slices were then digitally stacked along the z axis to yield the final 3D morphology.

To simulate the electric potential and field distributions, a 2D finite-difference method (FDM) was implemented using a successive over-relaxation (SOR) scheme. The simulation domain spanned $20\text{ nm} \times 15\text{ nm}$ with a spatial resolution of 0.02 nm . The lower 10 nm of the domain was assigned a relative permittivity of $\epsilon = 50$ to represent the TiO_2 layer, while the remaining region was treated as air ($\epsilon = 1$). The BE surface protrusion was modeled as a Gaussian profile with a height of 1.2 nm and a standard deviation of $\sigma = 0.8\text{ nm}$, where the latter was chosen to account for tip-induced lateral broadening inherent to C-AFM imaging: because the measured protrusion width reflects a convolution of the true surface geometry with the finite tip radius, a narrower width was used in the simulation to better approximate the intrinsic protrusion geometry.²⁶ The conductive AFM tip was approximated as a hemisphere of radius 20 nm positioned at the top of the TiO_2 layer, with a bias of $+5.5\text{ V}$ applied to the tip and 0 V to the BE. Convergence was determined when the maximum nodal update fell below a tolerance of 10^{-7} . The electric field components were subsequently obtained from the spatial derivatives of the converged potential. Applying $+5.5\text{ V}$ to the tip with the BE grounded produces the same field distribution as the experimental configuration (-5.5 V on the BE, tip grounded); an axisymmetric equivalent of this simulation, which accounts for the rotational symmetry of the conical filament, is provided in the [Supporting Information](#).

We acknowledge that the conductive AFM tip geometry differs from the planar electrodes used in practical RRAM devices. However, this configuration serves as a well-defined model system with two key advantages. First, the precise electric field localization at the tip apex enables controlled formation and spatial isolation of a single CF, which is essential for the tomographic reconstruction presented here. Second, the BE interface physics—including the field-enhancement effect of nanoscale surface protrusions and the resulting spatial confinement of oxygen vacancies—is independent of the TE geometry. The mechanistic insights derived from this model system are therefore relevant to understanding CF behavior in conventional planar-electrode devices.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.6c02423>.

Notes S1–S13 (FDM simulation, competition model, current-threshold sensitivity, tip convolution, measurement/sign convention, instrumentation and probe, force calibration, tip-wear monitoring, BE roughness, self-heating analysis, axisymmetric field model, protrusion field enhancement, and reproducibility across independent filaments), Figures S1–S8, and Table S1 ([PDF](#))

Sequential YZ cross-sectional current maps through the tomographic volume (Movie S1a) ([GIF](#))

Sequential XZ cross-sectional current maps through the tomographic volume (Movie S1b) ([GIF](#))

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Author Contributions

K.P. and S.H. conceived and designed the study, K.P. performed C-AFM measurements and tomography experiments, conducted the FDM simulations, analyzed and interpreted the data, and wrote the manuscript. C.G. assisted with the C-AFM measurements. S.P. prepared the TiO_2/Pt thin-film samples. S.-H.K.P. provided guidance on sample fabrication and revised the manuscript. S.H. supervised the project and critically revised the manuscript. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

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