

# Advanced Electrical and Thermal Scanning Probe Lithography for Next-Generation Nanodevice Fabrication

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## ABSTRACT

Scanning probe lithography (SPL) offers a variety of physical and chemical techniques for surface modification, enabling the fabrication of customized, functional nanoscale features. These methods provide unmatched resolution, repeatability, and operational simplicity compared to other lithographic techniques. This paper reviews recent advancements in SPL driven by electrical and thermal effects, highlighting how these approaches address the fabrication of functional nanostructures. These advancements pave the way for next-generation nanotechnological applications, including single-dopant atomic devices, memristors, metasurfaces, nanofluidic devices, and more.

**Keywords:** Scanning Probe Lithography, Nanodevices, Nanofabrication, Field Emission, Oxidation, Thermal

## 1. INTRODUCTION

Nanomanufacturing techniques have enabled the creation of small structures with unique properties arising from quantum, electromagnetic, and thermal effects. This has led to the emergence of a wide range of nanotechnological applications, including nanoelectronics, nanophotonics, and photovoltaics. In recent years, next-generation nanodevices—such as single-electron transistors, quantum bit (qubit) devices, photonic devices, memristors, and neuromorphic computing devices—have demonstrated higher performance and enhanced functionality. For example, quantum dots-based single-electron transistors (SETs) become the promising building blocks for beyond-CMOS devices<sup>1</sup>. Their fabrication requires sub-10 nm scaling<sup>2</sup>, which offers ultra-low-power operation and high sensitivity, making them promising for both nanoelectronics and quantum computing<sup>3,4</sup>. Beyond extreme size reduction, complex geometrical nanostructures play a vital role in advanced device functionality, particularly in photon manipulation, surface plasmon polaritons, and nanoparticle manipulation<sup>5</sup>. Engineered topographical features enable precise control of photons, or nanoparticles, enhancing their functionality in sensing, photodetection, and energy harvesting. In addition, two-dimensional materials (e.g., graphene, MoS<sub>2</sub>) and transition metal oxides (TMOs, e.g., VO<sub>2</sub>, TiO<sub>2</sub>) are key materials for next-generation nanodevices due to their unique electronic, optical, and physicochemical properties. The atomic-scale thickness, high carrier mobility (e.g., graphene), and direct bandgaps (e.g., MoS<sub>2</sub>) make them ideal for high-frequency transistors, flexible electronics, and optoelectronic devices, while the edge chemistry and interlayer coupling of 2D materials facilitate their use in quantum technologies, including quantum dots and single-photon sources. TMOs, such as VO<sub>2</sub>, exhibit metal-insulator phase transitions, enabling ultrafast switches and smart optical components. The tunability of TMOs through chemical doping or external fields supports applications in resistive memory (RRAM), neuromorphic computing, and catalysis. The precise fabrication of these functional nanostructures, along with the fine-tuning of their chemical and physical properties, is essential for their operation<sup>3,6</sup>.

However, as feature sizes continue to shrink, and geometry and material complexity increase, the successful realization of these devices imposes increasingly stringent demands on nanomanufacturing techniques. As many of these novel devices are still in development, the demand for efficient, cost-effective, and flexible ‘lab-to-fab’ nanofabrication techniques is increasing to facilitate the transition from research to large-scale manufacturing.

Current semiconductor-based nanofabrication approaches, including electron beam lithography (EBL), focused ion-beam lithography (FIBL), optical lithography, and nanoimprint lithography, are either expensive, slow or suffer from high defect rates due to multi-step processing for complex nanostructures<sup>7</sup>. For example, optical lithography and EBL/FIBL often rely on capital-intensive equipment with the cost ranging from \$1 million for an EBL/FIBL system to \$450 million for the latest extreme ultraviolet lithography (EUVL) machine<sup>8</sup>. EBL, and FIBL are slow, and limited in scalability, and suffers from proximity effect, sample damage and contamination due to the use of high-energy beams<sup>7,9</sup>. Nanoimprint lithography is usually performed based on expensive industrial-scale machines and often suffers from a high defect rate during mold

release <sup>10</sup>. In addition, nanoimprint lithography, EBL, and optical lithography could be inflexible in generating nanostructures with various shapes, as one type of tool (mask/stamp) can only make one type of structure.

Scanning probe lithography (SPL), developed from scanning probe microscopy (SPM) <sup>11,12</sup>, offers high precision and flexibility by directly modifying surfaces through a nanometer-size probe, enabling patterning at nanometric and atomic scales, but with low instrument cost and environmental requirement (no vacuum) <sup>13-15</sup>. SPL can be easily achieved through both electrical (bias-induced), and thermal processes. External energy sources as electrical field and thermal provide more controllable and versatile patterning capabilities than mechanical SPL approaches. These advanced SPL techniques can target different materials and geometries, making them valuable for next-generation device fabrication. While SPL faces scalability challenges for industrial mass production, it remains a powerful tool for in-lab prototyping of novel functional devices. This paper provides an overview of recent SPL applications, particularly on the widely used electrical and thermal SPL processes, emphasizing their role in creating innovative nanodevices <sup>16</sup>.

## 2. BASICS OF SPL

AFM operates on straightforward principles, utilizing a sharp tip with a nanometer-scale radius, mounted on a microscale cantilever, to scan a specimen's surface. When the tip approaches the sample, forces between the tip and the surface induce a response in the cantilever, causing deflection in static/contact mode or altering vibrational motion in dynamic/tapping (non-contact) mode <sup>11</sup>. Depending on the type of interaction between the tip and the surface, AFM is able to image and measure diverse physical properties such as topography, friction, charge distribution, work function, local magnetic fields, electronic spins, and thermal conductivity.

Beyond imaging, AFM has advanced into a tool for direct material manipulation at the nanoscale <sup>12</sup>. Nanometric sharp tips can function as a tool to modify material surfaces through electrical, thermal, mechanical, or chemical interactions, or a combination of these <sup>14</sup>. The obtained nanostructures can exhibit special nanotechnological functions based on different materials and geometries. The dual capabilities in imaging and nanometer-scale fabrication have enabled ultrahigh-resolution and imaging-integrated lithography, establishing AFM as a potent technique in nanotechnology research and applications <sup>17</sup>. SPL can be achieved through different ways <sup>14</sup>, including the mechanical, bias-induced, oxidation, field emission, thermal, and dip-pen <sup>18</sup>. Existing research has proven them the potential device fabrication capabilities <sup>19,20</sup>. The present paper will focus on three types of the existing modalities: field-emission SPL (FE-SPL), oxidation SPL (o-SPL), and thermal SPL (t-SPL).

## 3. DEVICE FABRICATION VIA SPL

### 3.1 FE-SPL

FE-SPL utilizes Fowler-Nordheim tunneling electrons from an ultra-sharp tip to induce resist exposure on a sample. The highly localized field emission current, generated by a strong non-uniform electric field ( $\sim 10^9$  V/m) within the tip-resist gap, enables direct, cost-effective nanolithography with low-energy electrons ( $<50$  eV) <sup>21</sup>. FE-SPL supports both positive-tone and negative-tone modes, allowing patterning on various polymeric resists, including calixarene <sup>22</sup>, polystyrene (PS) <sup>23</sup>, H-passivation layers <sup>24</sup>, and silk fibroin protein <sup>25</sup>. The operation of FE-SPL is illustrated in Figure 1. By optimizing parameters such as electron dose, bias voltage, polarity, scanning velocity, and tip shape, FE-SPL achieves sub-5 nm resolution. Unlike electron beam lithography (EBL), FE-SPL minimizes the proximity effect and is compatible with post-patterning processes such as cryogenic or reactive ion etching (RIE), facilitating pattern transfer onto functional materials for nanoelectronic devices, dielectric structures, and optical detectors <sup>26</sup>.

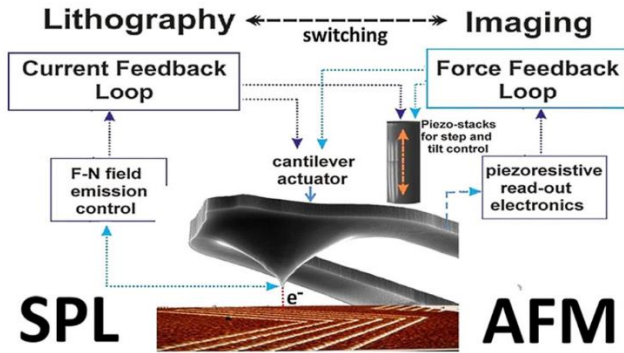


Figure 1. Schematic layout of the lithography system incorporating a current feedback loop for SPL and a force feedback loop for AFM imaging and probing<sup>27</sup>.

Recent advancements have integrated active cantilevers<sup>28–31</sup>, which enhance patterning resolution and allow direct, development-free exposure of calixarene using low-energy electrons. These cantilevers also enable high-resolution imaging before and after patterning, facilitating real-time inspection of samples. In addition, they allow precise overlay alignment and stitching of patterns, improving patterning accuracy. The compact, table-top FE-SPL system also offers reproducibility and controllability for beyond-CMOS device fabrication. This technology chain paves the way for routine and accessible manufacturing of single-electron devices. The general setup for tip-based nanolithography, incorporating two control feedback loops, is schematically illustrated in Figure 1.

Single-electron transistors (SETs) can operate at room temperature with charging energy  $E_c > k_B T = 25 \text{ meV}$  at  $T = 290 \text{ K}$ . Achieving this requires quantum dots smaller than 10 nm, which can be fabricated using FE-SPL. Conventional EBL-based approaches have demonstrated room-temperature SETs by defining  $50 \times 50 \text{ nm}$  point contacts, followed by heavy oxidation to reduce the unoxidized QD core to  $\sim 5 \text{ nm}$ . FE-SPL offers an alternative to EBL for SET fabrication, providing finer feature definition and reducing damage caused by high-energy electron beams<sup>16</sup>. Here, the FE-SPL was used to ablate a 15 nm thick calixarene layer, working as a positive resist. The low electron energies ( $< 100 \text{ eV}$ ) requires about 10 electrons per single calixarene molecule to induce a crosslinking event and a reaction scheme based on a direct ablation of resist material can be employed. The sensitivity was 80-times greater than a classical EBL, achieving line widths down to 10 nm for defining SET cores. The resist pattern was further transferred to the silicon layer using cryogenic  $\text{SF}_6/\text{O}_2$  plasma etching at  $120^\circ\text{C}$ , followed by thermal oxidation ( $850^\circ\text{C}$ , 10 min), producing quantum dots (QDs)  $< 5 \text{ nm}$  in diameter at the point-contact "neck" due to stress-limited oxidation. Ohmic contacts were defined using photolithography, Cr/Al evaporation, and lift-off. Electrical characterization confirmed single-electron charging effects, with observed Coulomb staircase behavior and a charging energy of 0.17 eV.

Rangelow et al.<sup>32</sup> further combined FE-SPL with single dopant lithography to fabricate dopant atom quantum dot SETs. Following the same procedure to pattern the  $\sim 10 \text{ nm}$  scale Si/SiO<sub>2</sub>/Si point-contact tunnel junction using FE-SPL, they used a hollow-tip active cantilevers to precisely implant dopants. This approach enabled quantum dot formation with atomic precision, ensuring room-temperature transistor operation for quantum and atomic-scale applications<sup>33</sup>.

Despite its advantages, FE-SPL faces challenges in writing speed and large-area scalability. Alternative tip materials such as gallium nitride, tungsten, platinum, and gold have been explored to address tip wear and contamination, improving long-term stability<sup>34–36</sup>. Integration with nanoimprint lithography can further enhance scalability for batch production, providing high-throughput pattern replication. In addition, mix-and-match lithography combining FE-SPL with optical lithography offers a viable strategy for improving throughput while maintaining nanoscale resolution.

Beyond resist exposure, FE-SPL has been investigated for tip-induced nanoscale 3D printing. Holz et al.<sup>37</sup> demonstrated field-emission scanning probe-induced deposition (TEBID), where low-energy electrons ( $< 100 \text{ eV}$ ) decompose precursor molecules, enabling localized 3D carbon nanostructure deposition within an SEM chamber. This approach allows fabrication of high-aspect-ratio nanowires and branched structures, showing potential for electrode fabrication, field emission devices, and functional nanoscale components.

### 3.2 Oxidation SPL

o-SPL is a process that occurs within a simple nanoelectrochemical cell, with a schematic illustrated in Figure 2, where the AFM tip acts as the cathode, the water bridge between the tip and substrate functions as the electrolyte, and the substrate serves as the anode<sup>38–40</sup>. Unlike conventional anodic oxidation—which employs roughly Avogadro’s number ( $\sim 6 \times 10^{23}$ ) of electrolyte molecules—tip-induced local anodic oxidation (LAO) relies on approximately  $1 \times 10^4$  to  $1 \times 10^5$  water molecules to oxidize material surfaces<sup>41</sup>. The water bridge forms via mechanical contact or electrical field application, particularly in dynamic modes. When a few volts are applied, the resulting electric field ( $1 \times 10^9$  to  $1 \times 10^{10}$  V/m) ionizes water molecules, generating oxidative species (such as  $\text{OH}^-$  and  $\text{O}^-$ ) that locally oxidize the material surfaces. The high aspect ratio of the AFM tip further concentrates this field, enhancing oxidation efficiency<sup>41</sup>.

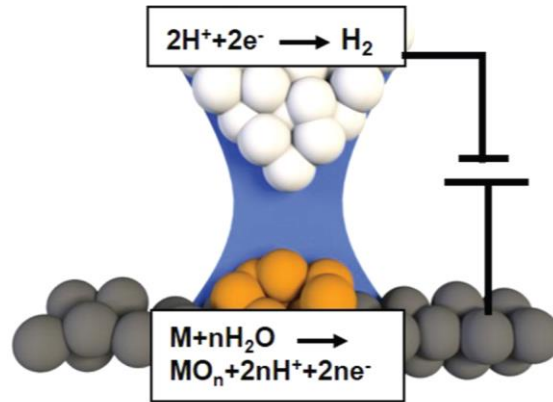


Figure 2. General schematic of nanopatterning using o-SPL<sup>40</sup>.

Since its introduction<sup>42</sup>, o-SPL has been widely applied in etch masks, biomolecule binding sites, and conjugated materials<sup>43</sup>. More recently, it has played a key role in next-generation nanodevices, offering flexible, precise control over nanoscale regions and enabling tunable chemical and physical properties of novel materials.

Similar to FE-SPL, o-SPL, combined with chemical etching, can also create surface localized nanostructures by performing LAO on thin-film metals or semiconductors, allowing for the flexible definition of functional device regions or channels. Herranz et al.<sup>44</sup> demonstrated the fabrication of InAs quantum dots within nanoholes created by LAO and selective wet etching on an epitaxial GaAs(100) substrate. These InAs quantum dot layers served as seed layers for stacking optically active, site-controlled InAs QDs. Similarly, using a scanning probe, o-SPL can pattern nanowires and define Coulomb blockage on metal or semiconductor surfaces, with smallest feature size approaching sub-10 nm. These structures have been utilized in various applications, including field-effect transistors<sup>45</sup>, and single molecule detection<sup>46,47</sup>.

Two-dimensional (2D) materials have emerged as a key building blocks for next-generation nanoelectronics, photonics, and quantum technologies due to their atomically thin structure and unique electronic properties. However, the performance of 2D nanodevices depends strongly on edge quality and chemical composition<sup>48</sup>. Conventional high-energy beam-based lithography techniques can induce substrate damage, necessitating alternative approaches for precise edge control and defect minimization. As a resist-free and direct-write technique, O-SPL enables clean and precise patterning of 2D materials, achieving extremely narrow nanoconstrictions on graphene and  $\text{MoS}_2$  ( $\sim 20$  nm)<sup>49–52</sup>. These nanostructures exhibit quantum confinement effects, enabling the formation of single quantum dots, Schottky junctions, and in-plane transistors<sup>53–56</sup>.

Transition metal oxides (TMO), such as  $\text{TiO}_x$  and  $\text{VO}_x$ , play a crucial role in next-generation nanodevices due to their high information density, low energy consumption, and multilevel resistive switching capabilities. These materials enable applications in high-density memory, neuromorphic computing, and optoelectronics. O-SPL has been employed to fabricate forming-free TMO devices, simplifying the manufacturing process while maintaining excellent electrical stability and precise morphological control. Avilov et al.<sup>57</sup> developed and simulated titanium oxide nanostructures through o-SPL for artificial synaptic devices, examining the impact of anodization on oxygen vacancy distribution and oxide formation ( $\text{TiO}$ ,  $\text{Ti}_2\text{O}_3$ , and  $\text{TiO}_2$ ). Their electrochemical  $\text{TiO}$ -based prototypes demonstrated resistive switching between high (HRS)

and low (LRS) resistance states over 100,000 cycles, with LRS stability for 10,000 seconds. Multilevel switching was achieved by varying the set voltage, highlighting TiO nanostructures' potential for neuroelectronics and neural networks. Tominov et al.<sup>58</sup> investigated gallium oxide nanostructures fabricated via o-SPL, demonstrating stable, forming-free resistive switching with an HRS of  $11.7 \pm 1.6 \text{ G}\Omega$  and an LRS of  $2.3 \pm 0.8 \text{ G}\Omega$ . The results confirm the feasibility of LAO-based memristive devices for neuromorphic computing. Tominov et al.<sup>59</sup> explored electrochemical titanium oxide structures with thickness-dependent resistive switching. They reported a resistance ratio (RHRS/RLRS) decreasing from 242.8 to 10.6 as thickness increased, with stable bipolar switching and no electroforming required. XPS analysis confirmed the presence of  $\text{TiO}_2$ ,  $\text{Ti}_2\text{O}_3$ , and  $\text{TiO}$ , demonstrating the suitability of o-SPL for high-performance, forming-free memory devices. Zhang et al.<sup>60</sup> developed a  $\text{VO}_2$ -based tunable metasurface using electric-field SPL, achieving sub-100 nm precision via negative bias AFM etching and sonication. Their design, incorporating  $\text{VO}_2$ -gold nanoblocks, enabled tunable reflectivity in the near- and mid-infrared range, showcasing high-resolution, maskless patterning for advanced nanophotonic and optoelectronic applications.

### 3.3 Thermal SPL

t-SPL is a high-resolution, maskless nanolithography technique that enables direct patterning with lateral resolution below 10 nm<sup>61-63</sup>. It primarily works by heating a nanoscale probe tip (Figure 3) to near the sublimation temperature of a polymer resist, such as PMMA, polyphthalaldehyde (PPA), causing localized resist removal<sup>64,65</sup>. The resulting resist pattern can be used as a mask for substrate pattern transfer. Beyond resist sublimation, t-SPL facilitates thermally activated surface modifications, including chemical functionalization, magnetic phase transitions, and structural phase transformations through localized atomic rearrangements [63–66]. Today, t-SPL achieves sub-20 nm half-pitch patterning with features as small as 7 nm. It offers high-speed processing up to 20 mm/s while avoiding electron proximity effects and substrate damage. Integrated in situ imaging allows real-time monitoring and corrections, enabling markerless overlay and alignment on preexisting structures. In addition, t-SPL supports complex 3D patterning without the need for vacuum or masks, making it a highly versatile nanofabrication technique<sup>66-69</sup>.

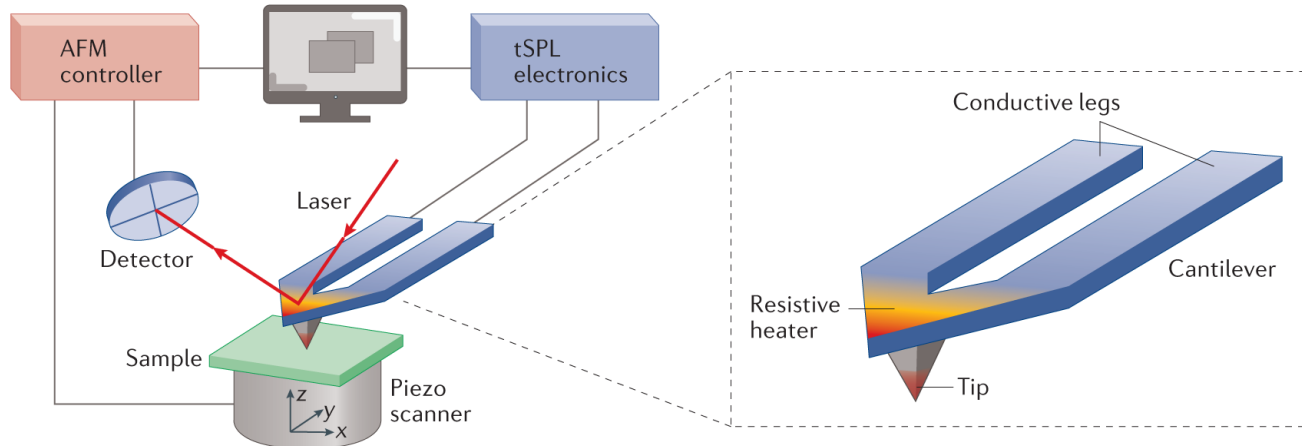


Figure 3. Schematic of t-SPL<sup>63</sup>.

A wide use of t-SPL is through locally modifying a thermally responsive resist layer using a heated probe, enabling the creation of nanoscale patterns with high precision and minimal substrate damage. Upon etching, these nanostructures can be transferred to the semiconductor surfaces to serve as functional components. Through this method, researchers have achieved nanodevices including 2D materials-based field-effect transistors<sup>70,71</sup>, nanowire-based quantum devices<sup>72</sup>, tellurium nanoribbon field-effect transistors<sup>73</sup>, high temperature superconducting Josephson junctions<sup>74</sup>, and silicon point-contact quantum-dot transistors<sup>75</sup>.

The ability of t-SPL to precisely control patterning depth has enabled its application in creating potential landscapes and complex 3D structures, which are challenging to achieve with conventional lithography techniques<sup>65</sup>. Skaug et al.<sup>76</sup> used t-SPL to shape the geometry of a nanofluidic slit. Combined with gentle shaking to induce a rocking Brownian motion, they were able to guide particles smaller than 100 nm along complicated paths. Lassaline et al.<sup>5</sup> demonstrated the use of t-SPL to generate smooth potential landscapes. The process involves using a heated silicon tip to locally sublimate a thermally sensitive resist deposited on a substrate. This technique allows for the creation of complex 3D structures and

gradient patterns with sub-100 nm feature sizes and nanometer precision. The technique was shown to be effective for creating optical Fourier surfaces, plasmonic band structures, and electronic potential landscapes.

The thermal energy delivered from the tip can also modify the materials, especially thin films, through inducing chemical reaction, crystallization, and local atom arrangement. Wei et al. <sup>77</sup> demonstrated a method for tuning the electrical properties of graphene oxide (GO) at the nanoscale using thermochemical nanolithography (TCNL) with a heated AFM tip. The technique allowed for the local reduction of GO, resulting in a significant increase in conductivity and the creation of nanoribbons as narrow as 12 nm. This method was proven to be clean, rapid, and reliable, with no observed tip wear or sample tearing, making it suitable for large-scale graphene electronics. Raghuraman et al. <sup>78</sup> introduced a technique to measure the kinetics of surface chemical reactions driven by localized heat and stress using an AFM. The study demonstrated the reduction of graphene oxide by varying the temperature and force applied by a heated AFM probe. The results revealed an activation energy of 0.55 eV and a first-order reaction, indicating an alternative reduction pathway compared to bulk processing methods. This technique can be extended to study the surface chemistry of other two-dimensional materials under various external stimuli. Hamann et al. <sup>79</sup> explored advancements in ultra-high-density phase-change storage using thermal recording techniques. They demonstrated erasable thermal phase-change recording at a storage density of 3.3 Tb inch<sup>-2</sup>, achieved by using a heated AFM tip to locally crystallize an amorphous chalcogenide film. They also introduced thin-film nanoheaters capable of generating hot spots smaller than 50 nm, showing potential for high-density data storage. The study further presented an all-thermal storage/memory concept, where a single heater is used for writing, erasing, and reading phase-change materials, highlighting the potential for overcoming the diffraction limit of optical recording and advancing non-volatile memory technologies. Albisetti et al. <sup>80</sup> demonstrated a method to create reconfigurable magnetic nanostructures using t-SPL. By locally heating a ferromagnetic layer coupled to an antiferromagnetic substrate in the presence of a magnetic field, they were able to control the direction and strength of magnetic anisotropy without altering the film's chemistry or topography. This method enables the creation of tunable spin wave devices and magnetic crystals, which are promising for future spintronic applications.

#### 4. CONCLUSIONS

High resolution lithographic techniques are typically linked with high capital investment in equipment, making them less accessible for rapid device development. As a result, alternative fast, direct, and low-cost prototyping techniques become essential in scientific research to accelerate innovation and exploration. SPL thus become an effective tool for on-demand prototyping next-generation nanodevices, particularly those with miniaturized and complex geometries. It is also well-suited for fabricating novel nanodevices based on emerging materials, such as 2D materials, oxide semiconductors, and phase change materials. With its ability to precisely define various intricate nanostructures, SPL plays a crucial role in advancing nanoelectronics and quantum technologies.

Despite its advantages, the application of SPL in next-generation nanodevice fabrication remains in its early stages. To achieve sustainable and scalable nanofabrication, several challenges must be addressed, including precise and reproducible pattern generation, high-speed and durable probe tips, and improved throughput. To enhance the practicality of SPL, AI-based automation <sup>81</sup> and parallelized probe arrays <sup>68</sup> offer promising solutions for improving alignment, scalability, and reliability. The combination with nanoimprint lithography <sup>82</sup> enables the high-throughput replication of nanoscale features, addressing the limitations of serial processing. In addition, hybrid approaches integrating AFM-based lithography with nanoimprint, EBL, and photolithography provide scalable, ambient-compatible nanomanufacturing solutions. With continued advancements in speed, reliability, and parallelization, electrical and thermal SPL hold significant potential for driving the development of next generation of nanophotonic, quantum communication, and nanoelectronic devices.

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#### REFERENCES

- [1] Durrani, Z., Jones, M., Kaestner, M., Hofer, M., Guliyev, E., Ahmad, A., Ivanov, T., Zoellner, J.-P. and Rangelow, I. W., "Scanning probe lithography approach for beyond CMOS devices," presented at SPIE

- Advanced Lithography, 26 March 2013, San Jose, California, USA, 868017.
- [2] Durrani, Z. A. K., “Coulomb blockade, single-electron transistors and circuits in silicon,” *Phys. E Low-Dimens. Syst. Nanostructures* **17**, 572–578 (2003).
  - [3] Morton, J. J. L., McCamey, D. R., Eriksson, M. A. and Lyon, S. A., “Embracing the quantum limit in silicon computing,” *Nature* **479**(7373), 345–353 (2011).
  - [4] Zwanenburg, F. A., “Silicon quantum electronics,” *Rev. Mod. Phys.* **85**(3), 961–1019 (2013).
  - [5] Lassaline, N., “Generating smooth potential landscapes with thermal scanning-probe lithography,” *J. Phys. Mater.* **7**(1), 015008 (2023).
  - [6] Rangelow, I. W., Ahmad, A., Ivanov, T., Kaestner, M., Krivoschapkina, Y., Angelov, T., Lenk, S., Lenk, C., Ishchuk, V., Hofmann, M., Nechepurenko, D., Atanasov, I., Volland, B., Guliyev, E., Durrani, Z., Jones, M., Wang, C., Liu, D., Reum, A., et al., “Pattern-generation and pattern-transfer for single-digit nano devices,” *J. Vac. Sci. Technol. B* **34**(6), 06K202 (2016).
  - [7] Fang, F. Z., Zhang, X. D., Gao, W., Guo, Y. B., Byrne, G. and Hansen, H. N., “Nanomanufacturing—Perspective and applications,” *CIRP Ann.* **66**(2), 683–705 (2017).
  - [8] Wiley, B. J., Qin, D. and Xia, Y., “Nanofabrication at High Throughput and Low Cost,” *ACS Nano* **4**(7), 3554–3559 (2010).
  - [9] Fang, F. Z., Zhang, N., Guo, D. M., Ehmman, K., Cheung, B., Liu, K. and Yamamura, K., “Towards atomic and close-to-atomic scale manufacturing,” *Int. J. Extreme Manuf.* **1**(1), 012001 (2019).
  - [10] Wu, D., S. Rajput, N. and Luo, X. C., “Nanoimprint Lithography - the Past, the Present and the Future,” *Curr. Nanosci.* **12**(6), 712–724 (2016).
  - [11] Binnig, G., Quate, C. F. and Gerber, Ch., “Atomic Force Microscope,” *Phys. Rev. Lett.* **56**(9), 930–933 (1986).
  - [12] Samori, P., “Scanning probe microscopies beyond imaging,” *J. Mater. Chem.* **14**(9), 1353–1366 (2004).
  - [13] Gao, J., Luo, X., Fang, F. and Sun, J., “Fundamentals of atomic and close-to-atomic scale manufacturing: a review,” *Int. J. Extreme Manuf.* **4**(1), 012001 (2021).
  - [14] Fan, P., Gao, J., Mao, H., Geng, Y., Yan, Y., Wang, Y., Goel, S. and Luo, X., “Scanning Probe Lithography: State-of-the-Art and Future Perspectives,” *2, Micromachines* **13**(2), 228 (2022).
  - [15] Garcia, R., Knoll, A. W. and Riedo, E., “Advanced scanning probe lithography,” *Nat. Nanotechnol.* **9**(8), 577–587 (2014).
  - [16] Lenk, C., Krivoschapkina, Y., Hofmann, M., Lenk, S., Ivanov, T., Rangelow, I. W., Ahmad, A., Reum, A., Holz, M., Glinsner, T., Eibelhuber, M., Treiblmayr, D., Schamberger, B., Chouiki, M., Chan, B. T., el Otell, Z. and de Marneffe, J.-F., “High-throughput process chain for single electron transistor devices based on field-emission scanning probe lithography and Smart Nanoimprint lithography technology,” *J. Vac. Sci. Technol. B* **37**(2), 021603 (2019).
  - [17] Rosa, L. G. and Liang, J., “Atomic force microscope nanolithography: dip-pen, nanoshaving, nanografting, tapping mode, electrochemical and thermal nanolithography,” *48, J. Phys. Condens. Matter* **21**(48), 483001 (2009).
  - [18] Oswald, E., Palanisamy, K. and Kranz, C., “Nanoscale surface modification via scanning electrochemical probe microscopy,” *Curr. Opin. Electrochem.* **34**, 100965 (2022).
  - [19] Notargiacomo, A., Foglietti, V., Cianci, E., Capellini, G., Adami, M., Faraci, P., Evangelisti, F. and Nicolini, C., “Atomic force microscopy lithography as a nanodevice development technique,” **7**.
  - [20] Dagata, J. A., “Device Fabrication by Scanned Probe Oxidation,” *5242, Science* **270**(5242), 1625–1625 (1995).
  - [21] Rangelow, I. W., Ivanov, T., Sarov, Y., Schuh, A., Frank, A., Hartmann, H., Zöllner, J.-P., Olynick, D. L. and Kalchenko, V., “Nanoprobe maskless lithography,” presented at SPIE Advanced Lithography, 11 March 2010, San Jose, California, 76370V.
  - [22] Kaestner, M. and Rangelow, I. W., “Scanning probe lithography on calixarene towards single-digit nanometer fabrication,” **3, Int. J. Extreme Manuf.** **2**(3) (2020).
  - [23] Krivoschapkina, Y., Kaestner, M., Lenk, C., Lenk, S. and Rangelow, I. W., “Low-energy electron exposure of ultrathin polymer films with scanning probe lithography,” *Microelectron. Eng.* **177**, 78–86 (2017).
  - [24] Yianni, S. A., Hofmann, M., Schenk, A. K., Reuter, C., Rangelow, I. W. and Pakes, C. I., “Mask-less nanostructuring of hydrogen terminated diamond using localized field emission scanning probe lithography (FE-SPL),” **9, Appl. Phys. Lett.** **120**(9), 093503 (2022).
  - [25] Bicer, M., Kumar, B. G., Melikov, R., Dogru, I. B., Sadeghi, S., Rangelow, I. W., Alaca, B. E. and Nizamoglu, S., “Silk as a biodegradable resist for field-emission scanning probe lithography,” **43, Nanotechnology** **31**(43), 435303 (2020).

- [26] Rangelow, I. W., Lenk, C., Hofmann, M., Ivanov, T., Lenk, S., Guliyev, E., Kaestner, M., Aydogan, C., Bicer, M., Alaca, B. E., Ates, O., Torun, H., Yalcinkaya, A. D., Ahmad, A., Reum, A. and Holz, M., “Single nano-digit and closed-loop scanning probe lithography for manufacturing of electronic and optical nanodevices,” *Nanophotonics Australas.* 2017 **10456**, 58–67, SPIE (2018).
- [27] Kaestner, M., Ivanov, T., Schuh, A., Ahmad, A., Angelov, T., Krivoschapkina, Y., Budden, M., Hofer, M., Lenk, S., Zoellner, J.-P., Rangelow, I. W., Reum, A., Guliyev, E., Holz, M. and Nikolov, N., “Scanning probes in nanostructure fabrication,” *J. Vac. Sci. Technol. B* **32**(6), 06F101 (2014).
- [28] Stricklin, I., Gotszalk, T., Behzadirad, M., Manske, E., Kissinger, T., Rangelow, I. W. and Busani, T. L., “Multipurpose active scanning probe cantilevers for near-field spectroscopy, scanning tunnel imaging, and atomic-resolution lithography,” *J. Vac. Sci. Technol. B* **41**(4), 042601 (2023).
- [29] Behzadirad, M., Rishinaramangalam, A. K., Feezell, D., Busani, T., Reuter, C., Reum, A., Holz, M., Gotszalk, T., Mechold, S., Hofmann, M., Ahmad, A., Ivanov, T. and Rangelow, I. W., “Field emission scanning probe lithography with GaN nanowires on active cantilevers,” *J. Vac. Sci. Technol. B* **38**(3), 032806 (2020).
- [30] Xia, F., Rangelow, I. W. and Youcef-Toumi, K., [Active Probe Atomic Force Microscopy: A Practical Guide on Precision Instrumentation], Springer International Publishing, Cham (2024).
- [31] Rangelow, I. W., Ivanov, T., Ahmad, A., Kaestner, M., Lenk, C., Bozchalooi, I. S., Xia, F., Youcef-Toumi, K., Holz, M. and Reum, A., “Review Article: Active scanning probes: A versatile toolkit for fast imaging and emerging nanofabrication,” *J. Vac. Sci. Technol. B* **35**(6), 06G101 (2017).
- [32] Rangelow, I. W., Schenkel, T., Persaud, A., Durrani, Z., Jones, M., Pietscher, H.-G., Dietrich, F., Tan, A. S., Stauffenberg, J., Manske, E., Froehlich, T. and Karpuzov, D., “Single Dopant Lithography for the Fabrication of Atomic-scale Devices and Quantum Systems,” 2024 31st Int. Conf. Mix. Des. Integr. Circuits Syst. Mix., 23–28 (2024).
- [33] Schofield, S. R., Fisher, A. J., Ginossar, E., Lyding, J. W., Silver, R. M., Fei, F., Nambodiri, P., Wyrick, J., Masteghin, M. G., Cox, D. C., Murdin, B. N., Clowes, S. K., Keizer, J. G., Simmons, M. Y., Stemp, H., Morello, A., Voisin, B., Rogge, S., Wolkow, R. A., et al., “Roadmap on Atomic-scale Semiconductor Devices,” *Nano Futur.* (2025).
- [34] Behzadirad, M., Mecholdt, S., Randall, J. N., Ballard, J. B., Owen, J., Rishinaramangalam, A. K., Reum, A., Gotszalk, T., Feezell, D. F., Rangelow, I. W. and Busani, T., “Advanced Scanning Probe Nanolithography Using GaN Nanowires,” *Nano Lett.* **21**(13), 5493–5499 (2021).
- [35] Stricklin, I., Gotszalk, T., Behzadirad, M., Manske, E., Kissinger, T., Rangelow, I. W. and Busani, T. L., “Multipurpose active scanning probe cantilevers for near-field spectroscopy, scanning tunnel imaging, and atomic-resolution lithography,” *J. Vac. Sci. Technol. B* **41**(4), 042601 (2023).
- [36] Lenk, C., Hofmann, M., Ivanov, T., Ahmad, A., Lenk, S., Rangelow, I. W., Reum, A., Reuter, C., Holz, M., Behzadirad, M., Rishinaramangalam, A. K., Feezell, D. and Busani, T., “Sharp GaN nanowires used as field emitter on active cantilevers for scanning probe lithography,” *J. Vac. Sci. Technol. B* **36**(6), 06JL04 (2018).
- [37] Holz, M., Hofmann, M., Allen, F. I., Weigel, C. and Strehle, S., “Tip-Induced 3D Printing on the Nanoscale with Field Emission Scanning Probes,” *Small* **21**(5), 2409035 (2025).
- [38] Gao, J., Luo, X., Xie, W., Qin, Y., Hasan, R. Md. M. and Fan, P., “Atomistic insights into bias-induced oxidation on passivated silicon surface through ReaxFF MD simulation,” *Appl. Surf. Sci.* **626**, 157253 (2023).
- [39] Luo, X., Gao, J., Xie, W., Hasan, R. Md. M. and Qin, Y., “Flexible single-step fabrication of programmable 3D nanostructures by pulse-modulated local anodic oxidation,” *CIRP Ann.* **72**(1), 177–180 (2023).
- [40] Ryu, Y. K. and Garcia, R., “Advanced oxidation scanning probe lithography,” *Nanotechnology* **28**(14), 142003 (2017).
- [41] Kurra, N., Reifengerger, R. G. and Kulkarni, G. U., “Nanocarbon-Scanning Probe Microscopy Synergy: Fundamental Aspects to Nanoscale Devices,” *ACS Appl. Mater. Interfaces* **6**(9), 6147–6163 (2014).
- [42] Dagata, J. A., Schneir, J., Harary, H. H., Evans, C. J., Postek, M. T. and Bennett, J., “Modification of hydrogen-passivated silicon by a scanning tunneling microscope operating in air,” *Appl. Phys. Lett.* **56**(20), 2001–2003 (1990).
- [43] Baumgärtel, T., Rehm, S., Würthner, F., von Borczyskowski, C. and Graaf, H., “Functional bisimide dyes bound via electrostatic interactions to oxide nanostructures generated by AFM lithography,” *Appl. Surf. Sci.* **318**, 51–58 (2014).
- [44] Herranz, J., González, L., Wewior, L., Alén, B., Fuster, D. and González, Y., “Study of Growth Parameters for Single InAs QD Formation on GaAs(001) Patterned Substrates by Local Oxidation Lithography,” *Cryst. Growth Des.* **15**(2), 666–672 (2015).

- [45] Espinosa, F., Uhlig, M. and Garcia, R., “Molecular Recognition by Silicon Nanowire Field-Effect Transistor and Single-Molecule Force Spectroscopy,” *1, Micromachines* **13**(1), 97 (2022).
- [46] Yusoh, S. N. and Yaacob, K. A., “Study on the Physical Properties of a SiNW Biosensor to the Sensitivity of DNA Detection,” *19, Materials* **14**(19), 5716 (2021).
- [47] Hutagalung, S. D., Lew, K. C. and Darsono, T., “Nanoscale Patterning by AFM Lithography and its Application on the Fabrication of Silicon Nanowire Devices.”
- [48] Li, X., Wang, X., Zhang, L., Lee, S. and Dai, H., “Chemically Derived, Ultrasoft Graphene Nanoribbon Semiconductors,” *Science* **319**(5867), 1229–1232 (2008).
- [49] Pea, M., De Seta, M., Di Gaspare, L., Persichetti, L., Scaparro, A. M., Miseikis, V., Coletti, C. and Notargiacomo, A., “Submicron Size Schottky Junctions on As-Grown Monolayer Epitaxial Graphene on Ge(100): A Low-Invasive Scanned-Probe-Based Study,” *38, ACS Appl. Mater. Interfaces* **11**(38), 35079–35087 (2019).
- [50] Rienstra, R. W., Sultana, N., Shih, E.-M., Stocker, E., Watanabe, K., Taniguchi, T., Richter, C. A., Stroschio, J., Zhitenev, N. and Ghahari, F., “Electron transport in bilayer graphene nano constrictions patterned using AFM nanolithography,” arXiv:2412.08758 (2024).
- [51] Neubeck, S., Ponomarenko, L. A., Freitag, F., Giesbers, A. J. M., Zeitler, U., Morozov, S. V., Blake, P., Geim, A. K. and Novoselov, K. S., “From One Electron to One Hole: Quasiparticle Counting in Graphene Quantum Dots Determined by Electrochemical and Plasma Etching,” *Small* **6**(14), 1469–1473 (2010).
- [52] Cohen, L. A., Samuelson, N. L., Wang, T., Klocke, K., Reeves, C. C., Taniguchi, T., Watanabe, K., Vijay, S., Zaletel, M. P. and Young, A. F., “Nanoscale electrostatic control in ultraclean van der Waals heterostructures by local anodic oxidation of graphite gates,” *10, Nat. Phys.* **19**(10), 1502–1508 (2023).
- [53] Ryu, Y. K., Dago, A. I., He, Y., Espinosa, F. M., López-Elvira, E., Munuera, C. and Garcia, R., “Sub-10 nm patterning of few-layer MoS<sub>2</sub> and MoSe<sub>2</sub> nanoelectronic devices by oxidation scanning probe lithography,” *Appl. Surf. Sci.* **539**, 148231 (2021).
- [54] Espinosa, F. M., Ryu, Y. K., Marinov, K., Dumcenco, D., Kis, A. and Garcia, R., “Direct fabrication of thin layer MoS<sub>2</sub> field-effect nanoscale transistors by oxidation scanning probe lithography,” *10, Appl. Phys. Lett.* **106**(10), 103503 (2015).
- [55] Dago, A. I., Ryu, Y. K., Palomares, F. J. and Garcia, R., “Direct Patterning of p-Type-Doped Few-layer WSe<sub>2</sub> Nanoelectronic Devices by Oxidation Scanning Probe Lithography,” *46, ACS Appl. Mater. Interfaces* **10**(46), 40054–40061 (2018).
- [56] Lee, D. H., Kim, C. K., Lee, J.-H., Chung, H.-J. and Park, B. H., “Fabricating in-plane transistor and memory using atomic force microscope lithography towards graphene system on chip,” *Carbon* **96**, 223–228 (2016).
- [57] Avilov, V. I., Tominov, R. V., Vakulov, Z. E., Zhavoronkov, L. G. and Smirnov, V. A., “Titanium oxide artificial synaptic device: Nanostructure modeling and synthesis, memristive cross-bar fabrication, and resistive switching investigation,” *7, Nano Res.* **16**(7), 10222–10233 (2023).
- [58] Tominov, R. V., Polupanov, N. A., Avilov, V. I., Solodovnik, M. S., Parshina, N. V., Smirnov, V. A. and Ageev, O. A., “Investigation of resistive switching in gallium oxide nanostructures formed by local anodic oxidation,” *1, J. Phys. Conf. Ser.* **1410**(1), 012233 (2019).
- [59] Tominov, R., Avilov, V., Vakulov, Z., Khakhulin, D., Ageev, O., Valov, I. and Smirnov, V., “Forming-Free Resistive Switching of Electrochemical Titanium Oxide Localized Nanostructures: Anodization, Chemical Composition, Nanoscale Size Effects, and Memristive Storage,” *8, Adv. Electron. Mater.* **8**(8), 2200215 (2022).
- [60] Zhang, W., Wu, X., Li, L., Zou, C. and Chen, Y., “Fabrication of a VO<sub>2</sub>-Based Tunable Metasurface by Electric-Field Scanning Probe Lithography with Precise Depth Control,” *ACS Appl. Mater. Interfaces* **15**(10), 13517–13525 (2023).
- [61] Garcia, R., Knoll, A. W. and Riedo, E., “Advanced scanning probe lithography,” *Nat. Nanotechnol.* **9**(8), 577–587 (2014).
- [62] Howell, S. T., Grushina, A., Holzner, F. and Brugger, J., “Thermal scanning probe lithography—a review,” *1, Microsyst. Nanoeng.* **6**(1), 21 (2020).
- [63] Albisetti, E., Calò, A., Zanut, A., Zheng, X., De Peppo, G. M. and Riedo, E., “Thermal scanning probe lithography,” *1, Nat. Rev. Methods Primer* **2**(1), 32 (2022).
- [64] Knoll, A. W., Pires, D., Coulembier, O., Dubois, P., Hedrick, J. L., Frommer, J. and Duerig, U., “Probe-Based 3-D Nanolithography Using Self-Amplified Depolymerization Polymers,” *Adv. Mater.* **22**(31), 3361–3365 (2010).

- [65] Pires, D., Hedrick, J. L., De Silva, A., Frommer, J., Gotsmann, B., Wolf, H., Despont, M., Duerig, U. and Knoll, A. W., “Nanoscale Three-Dimensional Patterning of Molecular Resists by Scanning Probes,” *Science* **328**(5979), 732–735 (2010).
- [66] Schwarz, C. M., Kuebler, S. M., Rivero-Baleine, C., Triplett, B., Kang, M., Altemose, Q., Blanco, C., Richardson, K. A., Du, Q., Deckoff-Jones, S., Hu, J., Zhang, Y., Pan, Y. and Ríos, C., “Structurally and morphologically engineered chalcogenide materials for optical and photonic devices,” *J. Opt. Microsyst.* **1**(01) (2021).
- [67] Carroll, K. M., Knoll, A. W., Wolf, H. and Duerig, U., “Explaining the Transition from Diffusion Limited to Reaction Limited Surface Assembly of Molecular Species through Spatial Variations,” *Langmuir* **34**(1), 73–80 (2018).
- [68] M. Carroll, K., Lu, X., Kim, S., Gao, Y., Kim, H.-J., Somnath, S., Polloni, L., Sordan, R., P. King, W., E. Curtis, J. and Riedo, E., “Parallelization of thermochemical nanolithography,” *Nanoscale* **6**(3), 1299–1304 (2014).
- [69] Albisetti, E., Petti, D., Pancaldi, M., Madami, M., Tacchi, S., Curtis, J., King, W. P., Papp, A., Csaba, G., Prod, W., Vavassori, P., Riedo, E. and Bertacco, R., “Nanopatterning reconfigurable magnetic landscapes via thermally assisted scanning probe lithography,” *Nat. Nanotechnol.* **11**(6), 545–551 (2016).
- [70] Conde-Rubio, A., Liu, X., Boero, G. and Brugger, J., “Edge-Contact MoS<sub>2</sub> Transistors Fabricated Using Thermal Scanning Probe Lithography,” *ACS Appl. Mater. Interfaces* **14**(37), 42328–42336 (2022).
- [71] Zheng, X., Calò, A., Albisetti, E., Liu, X., Alharbi, A. S. M., Arefe, G., Liu, X., Spieser, M., Yoo, W. J., Taniguchi, T., Watanabe, K., Aruta, C., Ciarrocchi, A., Kis, A., Lee, B. S., Lipson, M., Hone, J., Shahrjerdi, D. and Riedo, E., “Patterning metal contacts on monolayer MoS<sub>2</sub> with vanishing Schottky barriers using thermal nanolithography,” *1*, *Nat. Electron.* **2**(1), 17–25 (2019).
- [72] Shani, L., Chaaban, J., Nilson, A., Clerc, E., Menning, G., Riggert, C., Lueb, P., Rossi, M., Badawy, G., Bakkers, E. P. A. M. and Pribiag, V. S., “Thermal scanning probe and laser lithography for patterning nanowire based quantum devices,” *25*, *Nanotechnology* **35**(25), 255302 (2024).
- [73] Ying, H., Xu, M., Xu, X., Wen, L., Liu, Z., Wang, X., Zheng, X. and Huang, W., “Complete logic operations in an ambipolar tellurium homojunction via non-invasive scanning probe lithography,” *3*, *Device* **1**(3), 100069 (2023).
- [74] Duong, N. M. H., Berhane, A. M., Mitchell, D., Ullah, R., Zhang, T., Zhu, H., Du, J., Lam, S. K. H., Mitchell, E. E. and Bendavid, A., “Direct writing of high temperature superconducting Josephson junctions using a thermal scanning probe,” arXiv:2410.00372 (2024).
- [75] Rawlings, C., Ryu, Y. K., Rüegg, M., Lassaline, N., Schwemmer, C., Duerig, U., Knoll, A. W., Durrani, Z., Wang, C., Liu, D. and Jones, M. E., “Fast turnaround fabrication of silicon point-contact quantum-dot transistors using combined thermal scanning probe lithography and laser writing,” *Nanotechnology* **29**(50), 505302 (2018).
- [76] Skaug, M. J., Schwemmer, C., Fringes, S., Rawlings, C. D. and Knoll, A. W., “Nanofluidic rocking Brownian motors,” *6383*, *Science* **359**(6383), 1505–1508 (2018).
- [77] Wei, Z., Wang, D., Kim, S., Kim, S.-Y., Hu, Y., Yakes, M. K., Laracuenta, A. R., Dai, Z., Marder, S. R., Berger, C., King, W. P., Heer, W. A. de, Sheehan, P. E. and Riedo, E., “Nanoscale Tunable Reduction of Graphene Oxide for Graphene Electronics,” *Science* (2010).
- [78] Raghuraman, S., Elinski, M. B., Batteas, J. D. and Felts, J. R., “Driving Surface Chemistry at the Nanometer Scale Using Localized Heat and Stress,” *Nano Lett.* **17**(4), 2111–2117 (2017).
- [79] Hamann, H. F., O’Boyle, M., Martin, Y. C., Rooks, M. and Wickramasinghe, H. K., “Ultra-high-density phase-change storage and memory,” *Nat. Mater.* **5**(5), 383–387 (2006).
- [80] Albisetti, E., Petti, D., Pancaldi, M., Madami, M., Tacchi, S., Curtis, J., King, W. P., Papp, A., Csaba, G., Prod, W., Vavassori, P., Riedo, E. and Bertacco, R., “Nanopatterning reconfigurable magnetic landscapes via thermally assisted scanning probe lithography,” *6*, *Nat. Nanotechnol.* **11**(6), 545–551 (2016).
- [81] Degenhardt, J., Bounaim, M. W., Deng, N., Tutsch, R. and Dai, G., “A New Kind of Atomic Force Microscopy Scan Control Enabled by Artificial Intelligence: Concept for Achieving Tip and Sample Safety Through Asymmetric Control,” *1*, *Nanomanufacturing Metrol.* **7**(1), 11 (2024).
- [82] Barcelo, S. and Li, Z., “Nanoimprint lithography for nanodevice fabrication,” *Nano Converg.* **3**(1), 21 (2016).