

Special topic paper

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Molecular approach to semiconductors: a shift towards ecofriendly manufacturing and neuroinspired interfaces

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Abstract: Energy dissipation through physical downscaling towards more complex types of memory and logic devices, loss of ultrapure water and consumption of large amounts of (toxic) chemicals for wafer cleaning processes, as well as high thermal budget of solid-state synthesis and thin film growth of standard semiconductors including the use of rare earth elements – all this poses great challenges for semiconductor materials science and technology. Therefore, research and development of alternative methods for micro- and nanofabrication and chemical functionalization of a new type of resource- and energy-efficient semiconductors as the core component of every computer chip is crucial. One of the promising opportunities is the transformation of today's complementary metal-oxide-semiconductor (CMOS) electronics into ecofriendly and neuroinspired electronics driven by molecular design and multi-level switching mechanisms at room temperature. The sustainable chemical technology of electron transport and switching materials in semiconductor manufacturing and the development of devices with new unconventional nanophysics, improved performance, and augmented functionalities (beyond-CMOS and More-than-Moore) is becoming increasingly important in the context of a gradual transition to a future-oriented concept of Internet of Everything (IoE). In this article, we focus on the technological significance of semiconductor preparation from single-source (molecular) precursors and the prospect of functionalizing semiconductors using DNA origami nanotechnology and stimuli-responsive metal–oxygen cluster ions such as polyoxometalates (POMs). We also describe the advanced characterization of these qualified molecular systems by soft X-rays. We emphasize the technical relevance of using solution-based

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methods for the bottom-up preparation of novel and hybrid semiconductors as well as their challenging scalability and the compatibility of methods of molecular technology with lithography-based mass production. Our article aims to contribute to the achievement of the United Nations' Sustainable Development Goal 9 (Industry, Innovation and Infrastructure).

Keywords: 2D materials; biological computation; DNA origami; Global Conversation on Sustainability; molecular electronics; polyoxometalates; semiconductors; Sustainable Chemistry 2023.

Environmental impact and alternatives of electronic device manufacturing

The manufacturing of semiconductors for mobile phones, consumer electronics, PCs, information and communications technology (ICT) infrastructure, industry, and automobiles contributes significantly to greenhouse gas emissions (in particular, CO₂ and perfluorocarbons), water footprint, and chemical waste generation.^{1–3} The ever-evolving market for Internet of Thing (IoT) devices, high performance computing data centers, artificial intelligence (AI) algorithms, blockchain technologies (secure communication, cryptocurrencies), and automotive systems places burdens on energy and water resources and, thus, the climate conditions, especially in those regions with a high concentration of microelectronics industry and HPC/mining facilities.^{4–6} More than ever, basic and applied research must work hand in hand across the value chain to maximize the synergy with the Sustainable Development Goals of the United Nations, thus enabling green growth and sustainable development of current and next-generation electronic devices. It is necessary to reduce the general carbon footprint and its amount from the exponentially increasing demand for electricity from traditional digital computing and computational research, as well as consumption of ultrapure water during the silicon wafer cleaning process in the extremely energy-intensive semiconductor fabs.^{7–10}

It is obvious that the transition to a decarbonized 'electronic device' economy requires a high degree of innovation at every level of computer technology, that is chip manufacturing, hardware architecture design, mathematical models and software, and ICT infrastructure.¹¹ One very promising direction is the development of data-centric, neuroinspired computing from the bottom-up.^{12–16} Hereby, the human brain can be seen as an optimal antetype for energy efficiency, which, with a total power consumption of less than 20 W, impressively demonstrates that extreme parallelization and interconnection of rather slow-acting components – biological synapses – outperforms the extremely fast serial operating components that are still the backbone of the classical computing technology. Co-allocating memory and arithmetic logic (processing) units in a single computer memory cell (Fig. 1) is an important step towards that antetype, which promises to significantly improve performance and power efficiency of electronic devices. One of the central exploration zones to achieve this is the field of semiconductors, an area of material research that requires close collaboration between chemists, physicists, and engineers.

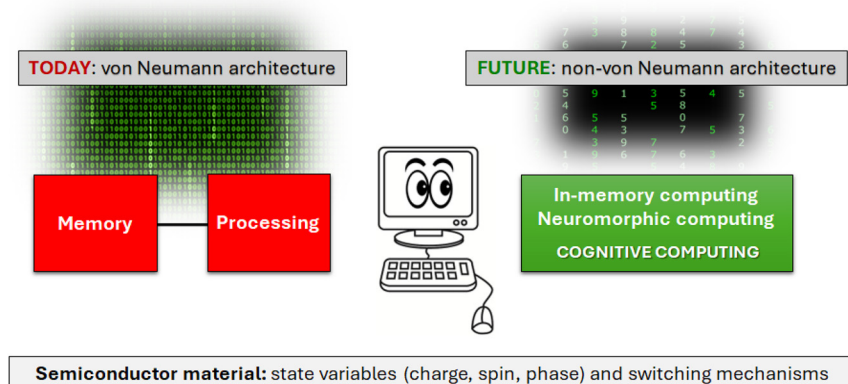


Fig. 1: Limits of today, challenges for tomorrow. The transition from classical digital computing (*left*) to resource-efficient and low-power, neuroinspired cognitive computing (*right*) based on environmentally friendly manufacturing of semiconductor materials.

Finding alternative, energy- and resource-efficient approaches to semiconductors is important for two reasons: (i) The micro- and nanofabrication of complementary metal-oxide semiconductor (CMOS) chips consumes billion kilowatt-hours of energy, utilizes tens of millions of tons of water, and produces million tons of carbon and thousands of tons of hazardous chemical waste annually.¹⁷ (ii) Almost all silicon-based computing using CMOS fabrication today is based on the archetypal von Neumann computer architecture, which suffers from considerable power consumption as well as high power dissipation rates due to heavy data transfer between memory and arithmetic logic unit.

Semiconductors and methods of their fabrication

As the semiconductor device dimensions are scaled down to a few nanometers and projected to reach the sub-nanometer range in 2030 and beyond, new challenges are posed to replace the traditionally used semiconductor materials.¹⁸ Two-dimensional (2D) materials such as graphene and transition metal dichalcogenides (TMDCs) are actually in the focus of research to overcome the limitations associated with further scaling down of silicon-based technologies. Device configurations based on 2D materials are expected to offer overarching opportunities for next-generation electronics, optics, and sensing related technologies.¹⁹ For the integration of new functional materials such as 2D materials into the device structures, there is ever growing interest for the development of sustainable and efficient processes. Using the top-down methods for patterning, the conventional photolithography will be limited with its resolution, while the patterning processes can be time consuming apart from the very high costs. In this context bottom-up approaches have garnered a lot of interest and there has been a paradigm shift in device technologies. It is striking to see how research in thin film technologies has enabled the development, fabrication, and integration of new semiconductor materials at the origin of devices used in our daily lives that range from electronics, optoelectronics, energy, etc.

Over the last decades there has been a surge in identifying new functional materials to be implemented in semiconductor devices to meet the demands of future technological innovations. Conventional semiconductors such as Si have been at the forefront of integrated circuits for several decades. Gradually new materials that include group III–V and group III nitrides, which provide several advantages over Si, have emerged with the continuous efforts in the development of epitaxy. As a result, more efficient, cheaper, and more ecofriendly devices have been realized for optoelectronic applications that include amongst others photovoltaic devices, wireless communications, radio frequency electronics, etc.

Thin films of semiconductors can be fabricated by a large variety of methods and their choice is frequently influenced by economic factors. Broadly the thin fabrication methods can be classified into three main categories depicted in Fig. 2: (i) chemical solution deposition (CSD), (ii) physical vapor deposition (PVD), and (iii) chemical vapor deposition (CVD). Although PVD yields device quality materials, it is often seen as the more expensive method due to the ultra-high vacuum requirements. For large-scale processing of semiconductor materials, the chemical gas phase methods are the front runners. CVD and atomic layer deposition (ALD) are widely acknowledged as powerful techniques for the deposition of high-quality layers for applications in microelectronics, optoelectronics, photovoltaics, sensors, etc. and most importantly they are industry compatible. CVD has dominated the semiconductor fabrication with thin film deposition for producing transistors with enhanced performances apart from producing other vital components such as epitaxial silicon, polycrystalline silicon, silicon nitride, III–V nitrides, etc. This technology is indispensable for creating components of today's electronics.²⁰ The importance of ALD as a true enabling nanotechnology is vastly growing providing opportunities for a wide range of application fields, both from a research and development perspective, and for high-volume manufacturing.^{21,22}

The semiconductor industry has been the main driver for the ALD technology in the last two decades. Especially, it has made a significant impact in the microelectronics sector. One of ALD's current frontiers is the

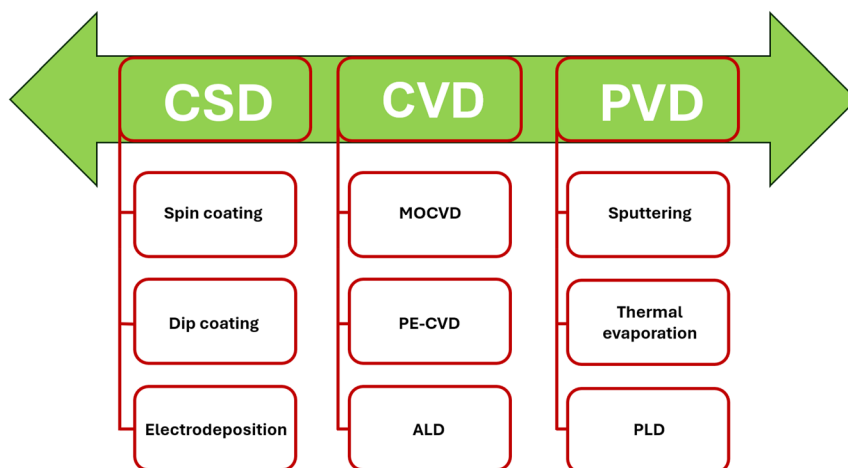


Fig. 2: An overview of thin film fabrication routes.

extension of this method to cover a wider range of materials. ALD processes also offer many advantages, like excellent step coverage with uniform thin film coating on nanosized and porous materials, high-aspect ratio structures, and 3D-architectures. Additionally, ALD is compatible in many manufacturing industries and hence offers the possibility of large-scale production. Apart from the temporal ALD process that is commonly used for growing thin films with atomic level precision, spatial ALD is an emerging variant where roll-to-roll deposition is achieved at atmospheric pressure with enhanced throughput at lower costs, scaling up to large area substrates, low material consumption especially for scarce materials, efficiency, and sustainability. The process is climate friendly that can be related to the reduction in energy consumption as no vacuum is needed, while the processing is performed at atmospheric pressure.²³ Area selective deposition (ASD) is emerging as a self-aligned process to overcome the limitations that are currently present in the fabrication of devices in the semiconductor industry.²⁴ A transition from top-down to bottom-up fabrication of microelectronic devices for continuous device miniaturization can prevent common issues such as the edge-placement error (EPE) in the layout of integrated circuits (ICs), or the use of highly expensive extreme ultra-violet (EUV) lithography on the smallest nodes. Local modification of substrate surface opens possibilities to achieve lateral control over film growth in addition to robust thickness during ALD process. Taking advantage of the surface chemistry differences between the growth and non-growth areas, the implementation of precursors will have a significant role to play for selective deposition. To achieve a selective deposition on desired materials, different strategies have been established. Most prominently, growth inhibition of a material on a non-growth area during ASD on patterned substrates can be realized by employing small molecule inhibitors (SMIs), self-assembled monolayers (SAMs), or surface modifications by plasma²⁵ (Fig. 3). ASD offers numerous advantages in nanoscale device fabrication such as reduction of the amount of required lithography steps, elimination of complicated etching processes, and minimization of expensive reagent usage.

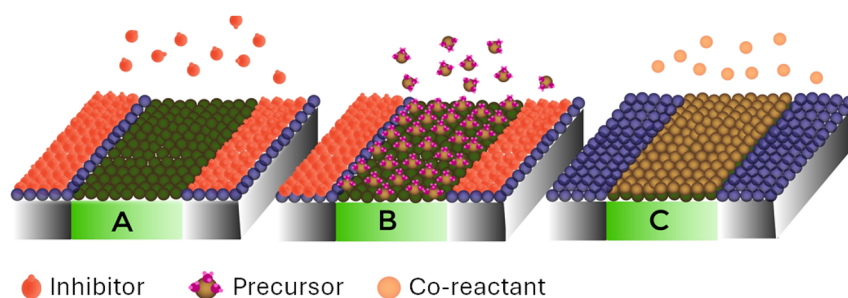


Fig. 3: Schematic of an area selective deposition (ASD) process.

Preparation of semiconductors from single-source molecular precursors

In pursuit of synthesizing semiconductor materials either as thin films or nanoparticles using chemical routes, wet chemistry and chemical gas-phase processes are predominantly used. For both cases precursors are employed either for solution deposition or CVD methods. Molecular precursors for spin, dip, or spray coating as well as for CVD and ALD have distinct advantages as the temperature of deposition can be lowered, enabling using of substrates that are thermally unstable such as polymers. Metalorganic precursors play a pivotal role in CVD and ALD of functional materials. Over the years, different generations of precursors have been employed starting from the metal halides, followed by organometallic precursors. There is a continuous demand in developing new or improved precursors to suit the demands of the material properties to be integrated into semiconductor device structures. As a representative example, for binary material systems such as GaN, trimethylgallium is the Ga source, while ammonia forms the nitrogen source. Epitaxial growth GaN is achieved at temperatures around 1,000 °C. On the other hand, it is possible to grow GaN at much lower temperatures using single source precursors (SSPs), where the molecule contains preformed Ga and N in the same molecule.²⁶ By introducing a ligand moiety that contains nitrogen bonded Ga, simplifies the materials processing route with the avoidance of the second precursor for nitrogen. 2D materials such as TMDCs flakes of high crystallinity are grown by CVD employing the sulfurization of metal oxides. Metalorganic precursors such as metal carbonyls in combination with H₂S yield crystalline MoS₂ or WS₂ films at high temperatures. Alternative precursors are sought after to avoid the toxic precursors. The application of nitrogen containing precursors of Mo and W in combination with elemental sulfur enables the formation of large-area MoS₂ and WS₂ thin films at moderate temperatures.^{27,28} The recent reports on SSPs for 2D materials such as TMDCs is one other approach.²⁹ These promising developments set the stage for new explorative molecular precursors for semiconducting materials that are targeted for future technological applications.

Single molecule precursors can also be utilized for the formation of mixed-metal semiconductor interfaces with a high level of atomic precision. For example, thin films of organically functionalized pentanuclear {Ce^{IV}₃Co^{III}₂} or dodecanuclear {Zr^{IV}₆Co^{II}₆} coordination clusters with a thermal stability of approx. 100 °C deposited on gold and silicon surfaces have shown promising potential for thermal transformation into pure and homogeneous mixed-metal oxide layers.³⁰ To convert the molecular precursor layers into organic-free metal oxide layers, not only thermal annealing processes can be used, but also photoinduction is an option. By changing the metal ratio of the precursors, the composition of the produced metal oxides can be adjusted in a very controlled manner without the typical problem of phase separation known from classical multi-stage CVD or PVD processes.

Molecular functionalization of semiconductors and its implication

The responsible electronics of tomorrow are expected to be based on materials and processes inspired by nature. The biologization of electronics towards more natural interfaces (enzymatic-like systems) would offer not only environmentally friendly devices and hardware, but also promises to grant access to biomimetic electron transport and switching systems. From this perspective, the coming years will be marked by more consolidating efforts to integrate the standard 'Si-CMOS' and heterogeneous 'Si-CMOS + 2D material' fabrication with molecular systems (Fig. 4).

There are many different compounds available in the basic molecular research market with potential applications in CMOS processes. Among the promising examples are DNA origami, peptide-based SAMs, and metal complexes capable of storing high charge capacity within their metal–oxygen and metal–metal bonds. Some of their representatives are discussed in detail in the following sections.

Unlike bulk materials with an extended solid structure, molecular compounds have discrete energy levels, which provides a universal lever for controlling the transfer of electronic charge within a device. Alike quantum dot (QD) qubits, charged and neutral molecular adsorbates can be combined with ferroelectric and resistive 2D

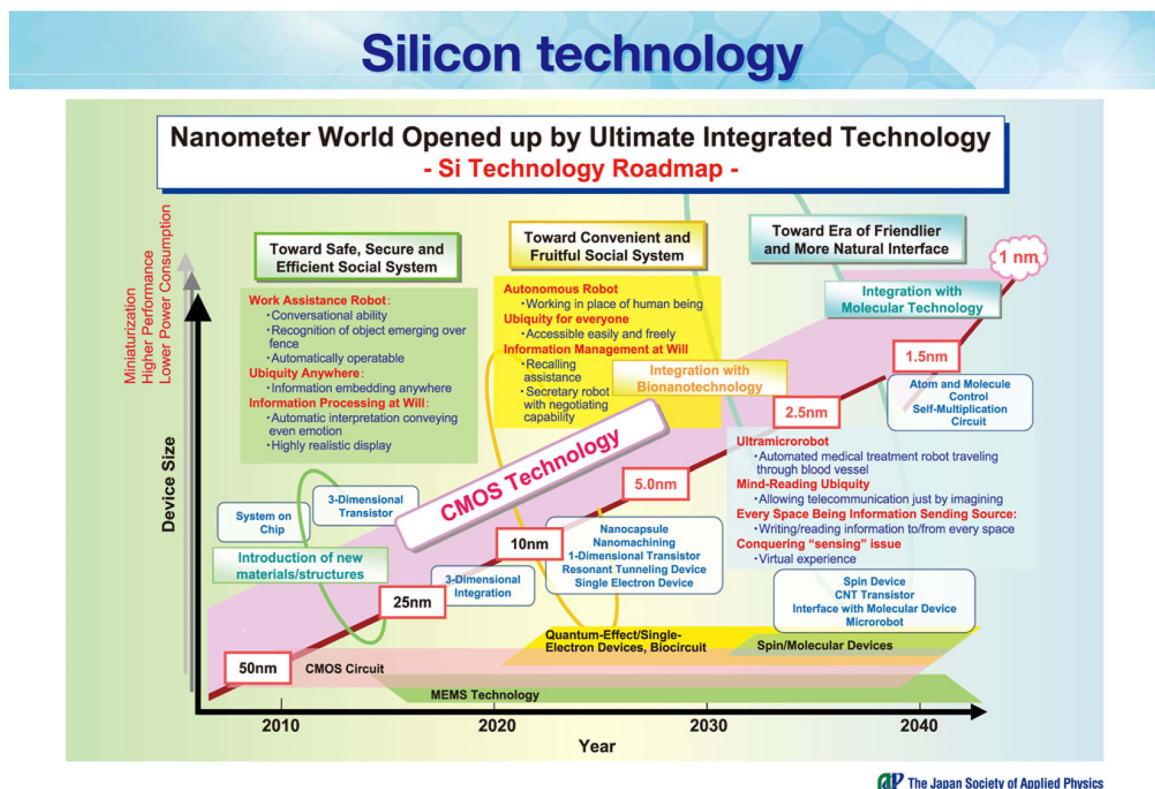


Fig. 4: Silicon technology roadmap. Copyright The Japan Society of Applied Physics.³¹

material surfaces, or used directly as active charge transport layers on silicon wafers. Stimuli-responsive molecules have the potential to add new chemical and physical functions that would positively impact a capacitance for multielectron transfer, modification of the electrical resistance, allow control of logic states and minimization of source-to-drain direct tunneling on sub-10 nm scales, and reduce parasitic leakage currents.

Challenges of integration of individual molecules and their assemblies into technical devices

In recent decades, microelectronic and semiconductor technologies have evolved more and more into the nanoscale. Both, bottom-up and top-down integration concepts of functional nanostructures and nanoparticles into micro- and nanofabricated electronic components have become critical issues that affect decisively the performance of smart electronic systems in various application areas such as environmental sensing and energy harvesting.

Nowadays, functional nanostructures and nanoparticles of different shapes, sizes, morphologies as well as materials are used in various technical and industrial applications, medicine, pharmacy, and biology. Nanoparticles (NPs) are also widely used in electronics, power engineering, ecology to solve environmental problems, development of new materials with unexpected properties, and many other fields.

In this respect, a complex engineering challenge is the integration of functional nanostructures such as molecular components (e.g. self-assembled molecular monolayers or entities of deoxyribonucleic acid, DNA), organic or inorganic nanoparticles (metal, semiconductor, colloidal materials) or similar building blocks into nanolithographically fabricated environments. The respective manufacturing paradigms go beyond current CMOS- and microelectromechanical system (MEMS)-related fabrication schemes. It has become necessary to

elaborate scale-bridging and hybrid manufacturing schemes that combine established top-down manufacturing techniques for microelectronic devices, including aspects of nanotechnology, with bottom-up nanoassembly of molecules on unstructured and pre-structured substrates. Microsystems technology based on the combination of conventional semiconductor processing with biotechnological approaches has thus become an emerging field with manifold applications, especially in the field of sensor technology.

From a technological perspective, top-down lithographic nanostructuring technologies and bottom-up nanostructuring technologies are the basis for novel device concepts. A key strategy for miniaturized energy-efficient electronic devices is the integration of structured entities of functional nanoscopic building blocks into pre-structured substrate materials that come in the dimensions of semiconductor wafers,³² chips³³ or – most recently, targeting applications of the IoT – hybrid or modular chips comprising a number of chiplets.³⁴

In classical semiconductor technology, the predictive miniaturization along Moore's law classically came along with the availability of more and more precise subtractive fabrication techniques. With red brick walls regarding miniaturization in sight, ambassadors of the so-called More-than-Moore approach propagated an increase of functionalization by the integration of molecular-type material structures in prestructured substrates. The More-than-Moore approach started off the trajectory of Moore's law, with bottom-up (i.e. additive) approaches on top-down fabricated substrates as a vehicle to reach this target. At this level, top-down and bottom-up manufacturing concepts came to meet. The combination of both opens up opportunities for a modular device design and a combination of the economic targets of high-throughput and high-volume fabrication and their ultimate personalization achievably in a single manufacturing chain.³⁵

A key technology for bridging the gap between top-down and bottom-up is the DNA origami method. It enables a template-based self-organization with a high-precision arrangement of molecules and nano-objects whose dimensions and spacings are beyond the usual resolution limits of the top-down technology and are integrated into larger lithographically pre-structured binding sites in the substrate with 100 nm–10 nm by the biological DNA origami matrix. The diversity of this inter- and intramolecularly programmable matrix is not limited by the heterogeneity of the functionalities attached of bottom-up technology (QDs, NPs, CNTs, biomolecules, etc.),³⁶ but only by the size of the template itself. This is determined by the length of the cyclic DNA single strand (scaffold), which is folded by an excess of short oligonucleotides (staple strands) that are individually different in sequence to synthesise the DNA origami template. The underlying mechanism is that the two halves of a staple strand each have a complementary base sequence to two different regions of the scaffold, which pulls it together (Fig. 5(a)). The resulting templates reach sizes of around 100 nm² and are multi-faceted in both 2D and 3D shapes.³⁷ The binding of functionalities to the surface of the biological template is achieved by integrating an additional binding sequence into one or more staples, which are not part of the folding process and whereby can act as binding points protruding from the surface. The localization depends on the spacing of the individual bases in the base sequence of the DNA origami and reaches a resolution of 2–5 nm. However, for a locally controlled, selective surface integration, a modification of the substrate is required to generate the individual binding points based on a chemical contrast (Fig. 5(b)). This can be achieved by functional groups, which enable binding of the DNA origami (binding-attractive) or not (binding-resistant). Due to the polar nature of the DNA origami, polyelectrolytes or functional group terminated silanes are usually used as bond-attractive layers in addition to SiO₂.^{38,39} As bond-resistant layers, non-polar, chemically inert materials such as CF polymers or silanes are used as hydrophobic layers. Placement and orientation of individual DNA shapes on lithographically patterned surfaces.⁴⁰ The former are deposited in thin films and structured by nanolithography.⁴¹ The topography created by this nanostructuring can achieve additional alignment and selectivity of the DNA origami in the binding sites.

The combination of additive and subtractive techniques has allowed an efficient design of manufacturing concepts that improve the precision of local subtractive patterning of organic electronic transistor channels by a self-aligning laser ablation step relying on the capillary-driven additive deposition of a spectrally suitable absorbent on a pre-patterned substrate surface.⁴² However, a central challenge of additive techniques is a precise spatial localization of a (pre-)functional material on a planar substrate. As subtractive techniques such as photolithography and electron-beam lithography have reached a very high accuracy,^{35,43} a forced confinement of additive material deposits by large-area coating and subtractive pre-patterning of planar substrates has turned out a viable strategy. An example is the direct-write laser-induced patterning of silicon wafers supplied with

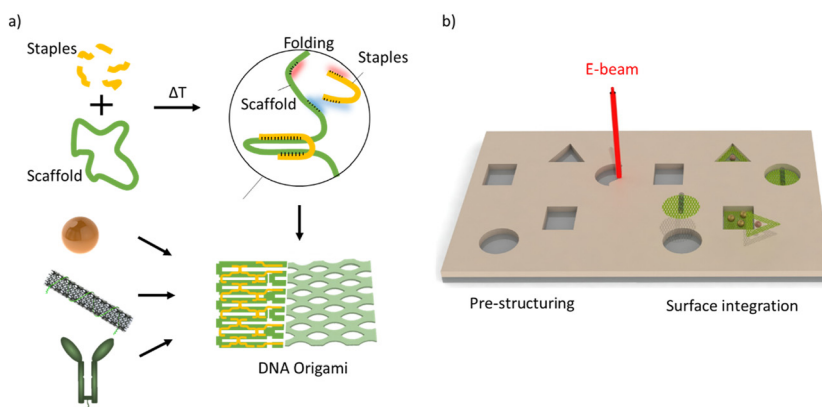


Fig. 5: DNA origami nanotechnology. (a) Synthesis mechanism of DNA origami based on long cyclic scaffold and short staple strands which folding the scaffold into individual programmable shapes with the possibility to attach heterogeneous functionalities as QDs, carbon nanotubes (CNTs), or biomolecules. (b) Controlled selective self-organisation of DNA origami hybrids (DNA origami with nanoobject) by pre-structured substrate.

large-area self-assembled molecular monolayers susceptible also to inkjet printing,⁴⁴ followed by vertical deposition and selective adsorption of nanoparticles in the voids of the pattern.⁴⁵

Additive manufacturing techniques rely on the spatial localization of a material that can be deposited but – especially in the context of electronic devices – does not necessarily include a functionality. Hence, these techniques often incur a functionality transformation of the applied (but not yet functional) material after its deposition. On the mesoscale, the field of printed electronics set a playground for additive techniques to such manufacturing concepts.⁴⁶ An example was the adaptation of the metallization approaches known from printed circuit board (PCB) technology using metal pastes to smaller dimensions employing inkjet printing of dissolved molecular complexes on the basis of metal-organic decomposition (MOD) solutions or molecularly stabilized nanoparticle dispersions.⁴⁷ In both cases, the spatial material deposition is followed by a tailored functionalization step (often called annealing, sintering, etc.) that is decisive for the functionality as electronic conductor.^{47,48}

Aside of the functionality of the individual component (such as the created metallic wiring), challenges that corroborate the device functionality regard the interface effects that occur in components fabricated serially by a number of additive techniques, which have to be understood in great detail to facilitate effective contacts and other aspects of the primary functionality.⁴⁹

Another important aspect in combining additive with subtractive techniques is the local alignment precision. The spatial localization and the geometry of the deposited material quantity is usually underlying uncertainties from individual deposit to individual deposit. Exemplified for printing, Belgardt et al. reported on the effects of a “dot touch” (a well studied phenomenon in graphical printing) during the printed deposition of SAMs on silicon oxide wafers and, hence, the surface coverage and the related achievable local surface energies.⁴⁴

Recent research activities at the example of point-like nanostructured attenuators⁵⁰ and nanostructured optical waveguides⁵¹ started to quantify the degree of deviation from order applicable e.g. for material deposits (but also to the results of subtractive methods) and correlate the morphology with conjectural quality parameters for optical devices such as spectral transmittivity. Demoly and André recently suggested to include considerations of a quantified disorder into the design of the targeted devices or components by programming or self-organization.⁵²

Aside of printing as a set of methods of direct materials deposition on wafer and chip level, flow chemistry has arrived in the world of microtechnologies as a method to facilitate chemical reactions at place in a scalable manner. According to Zhao et al., whilst developments in advanced manufacturing have achieved the miniaturization of semiconductor electronic devices to a near-atomic scale following continuously the ‘top-down’ construction method, huge technological challenges have been encountered with the exponentially increased cost and inevitably prominent quantum effects. Meeting these demands, ‘bottom-up’ methods aim to assemble

electronic devices from the molecular level. There is a rapid development of precise fabrication at a molecular scale, opening up horizons for a regulation at a quantum scale, and related applications of the basic electronic component of the ‘electrode–molecule–electrode junction’. The target is to control the quantum charge transport properties of the molecules and its interfaces with the environment,⁵³ which may go beyond the classical electrode-to-molecule-to-electrode sandwich structure but also functional entities that allow actuation or sensing. Traditionally, molecular functionalization on wafer level is bound to the gas phase, which allows a structured deposition of molecular features on wafer level in case of a presence of molecular binding sites achieved by pre-structuring.⁵⁴ Also wet chemistry approaches have been employed to pre-patterned substrates on chip level and wafer level to facilitate spatial localization of functionalizing nanoscopic building blocks on pre-fabricated transistor structures. Here, flow chemistry can be a technological approach for local molecular functionalization, and carbon-based nanomaterials have been found as an ideal ‘test bed’ for this task.⁵⁵ Hermann and co-workers have developed scalable functionalization approaches for carbon-nanotube field-effect transistors (CNT-FETs) made from single-walled carbon nanotubes (SWCNTs) localized between palladium electrodes on wafer, achieving a molecularly linked decoration of the SWCNTs in the transistor channels with preformed gold nanoparticles on chip level. They proved decoration via covalent thermally assisted [1,3]-dipolar cycloaddition of azide-terminated linkers to the SWCNTs⁵⁶ and non-covalent thermally assisted attachment of pyrene alkanthiol molecular linkers to the deposited SWCNTs,⁵⁷ of which the non-covalent functionalization protocol of the SWCNTs with gold nanoparticles could prove a successful operation of the achieved transistors as plasmonically enhanced spectrally selective optical sensors. Menna and co-workers have meanwhile developed and reported comparable technological concepts for a number of electronic and optoelectronic target devices comprising molecular moieties with carbon nanomaterials.^{58–60} Further approaches of wafer-level flow chemistry comprise organic functionalization of group IV semiconductor surfaces⁶¹ and functionalization of silica nanoparticles and native silicon oxide with boron-molecular precursors for efficient and predictive *p*-doping of silicon.⁶² Recent approaches related to flow chemistry regard wafer-level flow-modulation epitaxy facilitated e.g. by MOCVD of boron nitride on pre-structured sapphire substrates and mechanical exfoliation for low-power flexible electronic devices.⁶³ There are also examples of covalent grafting of polyoxometalate (POM) hybrids onto flat silicon oxide substrates aiming the functionalized POMs to be employed e.g. for molecular memory devices.^{64,65}

Micro- and nanostructured molecular layers for energy-efficient network-based biocomputing

Nanostructured layers hold significant importance for various applications, including microfluidic and microanalytical devices,^{66,67} biosensors,⁶⁸ and network-based biocomputation.^{69–71} Various additive and subtractive techniques, such as photolithography,^{72,73} dry-phase patterning,⁷⁴ inkjet printing with molecular⁴⁴ or nanoparticle-laden fluids,⁷⁵ stop-and-go microcontact printing,⁷⁶ dip-pen lithography,⁷⁷ layer-by-layer surface modification,⁷⁸ evaporation-induced self-assembly following liquid immersion and vertical deposition,⁷⁹ micro- and nanostructuring via laser ablation and desorption,⁴⁵ and force-controlled fluid deposition⁸⁰ can be applied to modify or exploit physicochemical properties, such as hyperlocal diffusion and wettability,^{81–83} charge, and optical characteristics (e.g., scattering, reflectance, and fluorescence).

In recent years, there has been growing interest in developing alternative parallel computing methods to address the limitations of conventional computing, solving problems sequentially. These methods aim to solve complex combinatorial problems more efficiently, which traditional computers struggle with due to the exponential increase in required calculations as the problem size grows. Among these innovative approaches are DNA computing⁸⁴ and quantum computing,⁸⁵ both of which have shown promise but face significant scalability challenges.

Network-based biocomputing (NBC) represents another parallel computing approach,⁸⁶ utilizing biological agents. A NBC system physically represents a mathematical problem graphically, which is then explored by

numerous autonomous agents, such as motile physical objects or cytoskeletal filaments driven by localized molecular motors,⁶⁹ to find a solution. These motile agents can be self-propelled microorganisms or passively guided cytoskeletal filaments,^{69,70,86–89} each with different operational modes and scaling parameters.⁹⁰

Large-scale fabrication of NBC systems leverages manufacturing techniques adapted from microelectronic device production. This includes incorporating aspects of nanotechnology and strategies for heterogeneous systems integration. The hardware for NBC is created using scale-bridging manufacturing schemes,⁷³ ensuring the approach is feasible for large-scale production and application. By adopting established methodologies from the microelectronics industry, NBC systems can be produced on a scale necessary to tackle complex combinatorial problems effectively.⁴³

The state-of-the-art in massively parallel alternate computation was set by quantum computing and DNA computing, which both have fundamental advantages over conventional computing. The largest nondeterministic polynomial-time complete (NP-complete) problem solved to date by quantum computing was Exact Cover with two sets, corresponding to the brute-force verification of four solutions.⁹¹ By contrast, experiments with DNA computing have been able to solve 3-SAT with a million potential candidate solutions.⁹² However, the continued progress of both technologies is unclear, with fundamental challenges such as de-coherence limiting the progress in quantum computation, and with the need for impractically large amounts of DNA plaguing efforts in DNA computing.⁹³

NBC is a parallel computing paradigm in which combinatorial problems are encoded into graphical networks realized by nanofabrication and then solved by parallel exploration of all paths through the networks using a large number of independent biological agents. As a key prerequisite for this approach, the networks must be designed such that all possible paths through the network correspond to all possible solutions to the combinatorial problem. Thus, exploration of these paths by autonomous biological agents then finds all possible solutions to the combinatorial problem. The largest NP-completed problem solved to date by protein filaments, that are propelled by molecular motors in a NBC device, was five-set Exact Cover, corresponding to the verification of 1,024 potential solutions.⁷¹ This results demonstrated a scalability of the NBC by a factor of 128, enabled by an improvement of network algorithm⁹⁴ and fabrication technology,⁷³ which reduced the error rates at junctions for biological agents as well as the search space. A further scale-up can be realized by narrower channels or improved junction geometries, which results in less junction errors and enables larger networks that encode larger problems. The integration of 3D pass junction geometries with bridges and tunnels^{95–97} would eliminate pass-junction errors altogether. These 3D micro-junctions, suitable biocomputational network devices, were designed and realized by 2PP of ORMOCER®, an organically modified hybrid material. The 3D junctions separated two perpendicularly aligned channels in the structure of bridge and a tunnel. Furthermore, simultaneous, spatially overlapping 2D filament transport driven by molecular motors along guide structures separated in the third dimension was demonstrated. These error-free junctions represent an essential advance for implementation in network designs to enable future upscaled biocomputing devices.

A further improvement of the networks can be achieved by integrating structured surfaces with switchable and rewritable patterns. One technique is the selective deposition of stimulating substances that form SAMs or polymer brushes. These structures enable local control of the topographical and physicochemical properties by externally manipulating the environmental conditions.⁹⁸ In this study, a pre-patterned surface was used to realize nanodimensional SAM-polymer structures. Therefore, an APDMES-SAM ((3-aminopropyl)-dimethylethoxysilane self-assembled monolayer), which consists of hydrophobic hydrocarbon chains was deposited in a 500 nm wide nanochannel of PMMA (AR-P679.04 photoresist from Allresist Berlin, Germany) on a SiO₂ surface. Subsequently, the SI-ATRP procedure⁹⁹ of PNIPAM (poly(N-isopropylacrylamide)) was used. The deposition of the PNIPAM functionalized surfaces was proven by contact angle measurements. The PNIPAM changes its structure above a LCST (lower critical solution temperature) and thus also the water contact angle. These measurements and further atomic force microscopy (AFM) investigations confirmed the successful selective grafting of the thermosensitive polymer PNIPAM in patterns produced by electron beam lithography, with feature sizes of 500 nm.⁹⁸ These results showed successful combination of the “top-down” lithography and patterning techniques with the “bottom-up” functionalization of the patterned structures using a SAM to enable the fabrication of switchable nanodimensional networks for energy efficient network-based biocomputing.

The energy efficiency of NBC, by solving complex mathematical problems, is due to the use of biomolecular motor proteins that transport biological agents, each only a few nanometres in diameter and a few micrometers in length through lithographically fabricated nanofluidic channel system. The motor proteins use adenosine-tri-phosphate (ATP) to propel the agents through the network, solving the complex problem in parallel. The energy required for the networks described in Nicolau *et al.* and Surendiran *et al.*^{69,71} was approximated to $\approx 4 \times 10^{-15}$ J/operation, which is orders of magnitude less than the 2 to 6×10^{-10} J/operation of an electronic computer.

Polyoxometalate redox capacitors and memristors

POMs are biogechemically relevant molecular compounds¹⁰⁰ and possess numerous useful properties both at the single molecule and thin film level, which can be exploited in semiconductor device technology. Vanadium-containing POMs (Fig. 6) can be switched as single molecules multiple times across their redox states at room temperature and low operating voltages (below 2 V), when the tip of a scanning tunneling microscope (STM) is used as the top electrode of a model resistive switching cell.^{101–103} Their anionic metal–oxygen cluster cores are able to communicate electronically with each other via counteranions or photoexcited states in solution and on

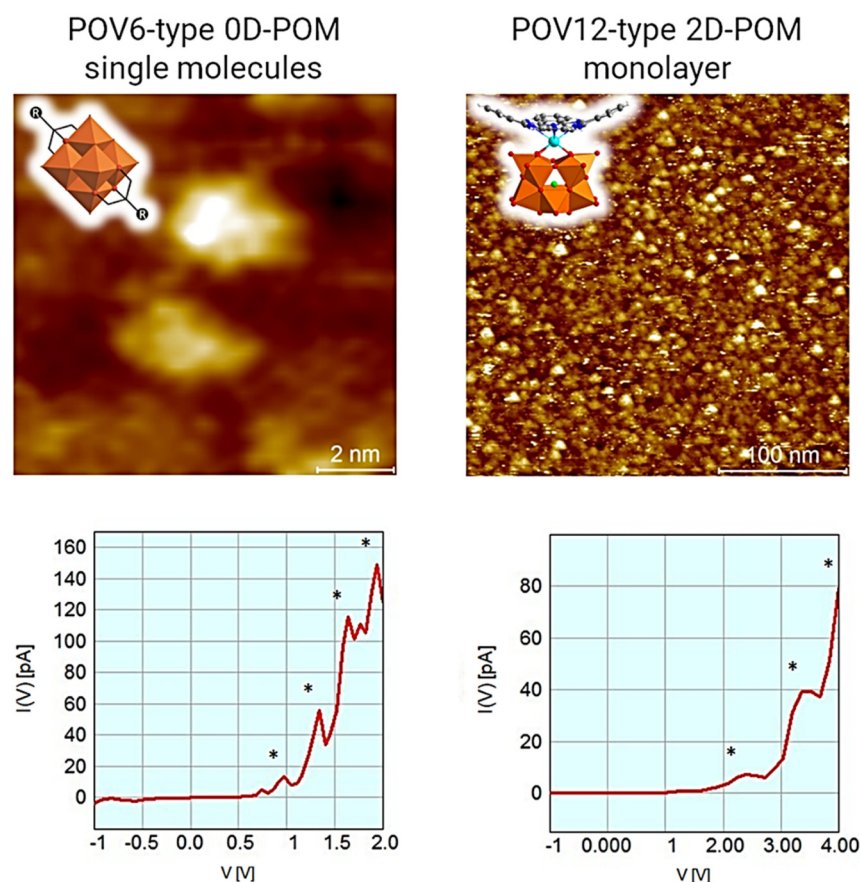


Fig. 6: Surface-confined polyoxovanadate (POV) structures as complementary molecular multilevel switches. *Left:* The tris(alkoxo)ligated Lindqvist-type hexavanadate $[V_6O_{13}((OCH_2)_3CR)_2]^{2-}$ with a compact octahedron-like structure composed of six V(3d) addenda atoms (POV6). The structurally exposed R endgroups can be effectively postfunctionalized with virtually any organic/inorganic moieties, which allows a tunable immobilization of the POV6 core on various substrate surfaces. *Right:* $[HV_{12}O_{32}Cl(LnPc)]^{3-}$ (POV12(LnPc), Ln = Sm^{III}–Er^{III}, Lu^{III} including Y^{III}). While POV6 preferentially adsorbs as single molecules with low (<2 V) voltage switching behavior, POV12(LnPc) tends to form a molecular monolayer with high (>2 V) voltage switching behavior.

surfaces.^{104,105} When covalently positioned on DNA origami, these POMs render it locally conducting on gold surfaces, demonstrating their extensive potential for the development of continuous conductance tunability (i.e., artificial synaptic functionality).¹⁰⁶ Overall, POMs with vanadium centers have proven to be excellent multilevel redox switches, exhibiting both capacitor and memristor behavior.¹⁰⁷ Furthermore, polyoxovanadates (POVs) functionalized with magnetic lanthanide phthalocyanine complexes have been shown to form conducting 2D monolayers on graphite surfaces, enabling multistate switching at voltages above 2 V at room temperature.¹⁰⁸ The combination of such responsive, different vanadium-containing POM structures with complementary electronic and adsorption properties opens up entirely new possibilities for the development of Boolean logic devices at the molecular level.

X-ray absorption spectroscopies for advanced characterization of polyoxometalates on surfaces

For the controlled integration of functional molecular compounds such as POMs into technical devices, a detailed knowledge of their electronic, magnetic, and structural properties is essential. One of the most powerful methods to study all these properties in an element-specific and in some cases even site-specific manner is X-ray absorption spectroscopy (XAS). For POMs, it allows the determination of intra- and intermolecular properties as well as the influence of substrates or surfaces in direct contact with deposited polyoxoanions, which is crucial for improving the desired topographical and electronic structural properties.

In particular, the X-ray absorption near-edge structure (XANES) is influenced by valence state, crystal fields, charge transfer, and delocalization/covalency. This is exemplarily shown in Fig. 7(a) and (b) for a POM structure hosting a central Fe(III) ion in the as-prepared state. By hydrogenation, the central ion changed to Fe(II), which is clearly visible by the shift in energy of the spectra. Spin and orbital magnetic moments have been determined from experimental data¹⁰⁹ by a sum-rule^{110–112} based analysis. Further analyses of spectral shape and magnetic response showed that the probed *d* electrons are less localized upon hydrogenation.¹⁰⁹ The change of valence state can easily be measured in other POMs as well including vanadium-containing ones.¹¹³

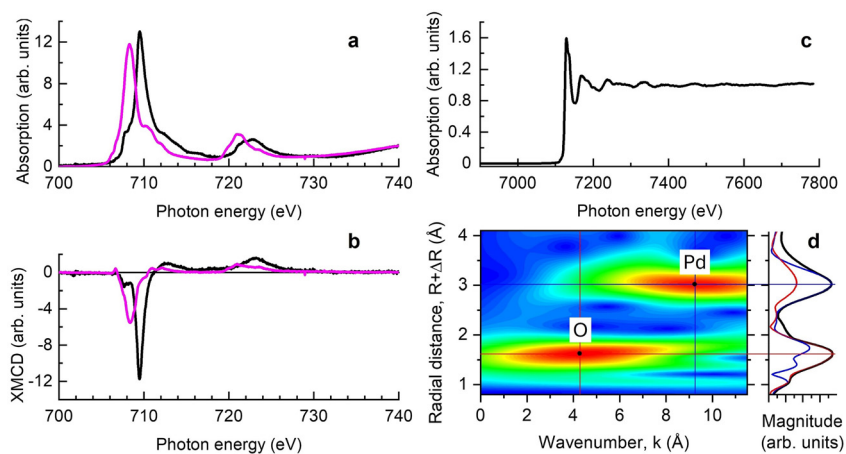


Fig. 7: X-ray absorption results for Fe-hosting POM. *Left:* X-ray absorption near-edge structure (XANES, a) and magnetic circular dichroism (XMCD, b) of an Fe-hosting POM in the as-prepared state (black lines) and after hydrogenation (magenta lines) measured at the Fe $L_{3,2}$ absorption edges. *Right:* Extended X-ray absorption fine structure measured for Fe-hosting POM at the Fe K absorption edge (c) and wavelet transform of extracted oscillations as function of photoelectron wavenumber and pseudo radial distance (d). Panels (a) and (b) show data from Ref.¹⁰⁹

For Pd in POMs, an unconventional diamagnetic state has been found¹¹⁴ that is facilitated by the strong spin–orbit coupling in $4d$ states. In this regard “unconventional” means that the spins of the eight d electrons in Pd(II) cancel out although formally occupying five and not four d orbitals. After this discovery, a magnetic phase diagram for Pd(II) in square-planar to sixfold coordination was calculated including paramagnetic, diamagnetic, and the unconventional diamagnetic state.¹¹⁴

Moreover, it has been shown that XANES, XMCD, and especially X-ray magnetic linear dichroism (XMLD) is sensitive to structural changes of POM induced by α -cyclodextrin molecules attached.^{115,116} A direct investigation of structural changes can be done by analyzing the extended X-ray absorption fine structure (EXAFS). EXAFS covers the energy range up to about 1 keV above the XANES region and originates from an interference effect between the outgoing photoelectron as a wave and backscattered waves from neighboring atoms. Frequency and amplitude of the EXAFS oscillations contain information about distance, number, and type of the atoms in vicinity of the absorbing element.

An example is shown in Fig. 7, again for Fe-hosting POMs. The EXAFS oscillations as a function of photoelectron wavenumber $\chi(k)$ are extracted after normalization, background subtraction, and finding the position of the absorption edge ($k = 0$). To suppress contributions of XANES and emphasize the exponentially damped EXAFS at higher k values, $\chi(k)$ is multiplied with k^n . The exponent depends on the system, data range and quality. The magnitude of the Fourier transform (FT) of the k^n -weighted $\chi(k)$ reflects the pseudo-radial distribution function (RDF) of backscattering atoms. Note that the distance values are usually shorter than the geometric distances in real space due to a phase shift. In a standard analysis procedure, experimental data in k and real space are compared with simulations based on multiple scattering theory. In nanoscale materials, where surface and interface effects play a crucial role, the number of parameters is often too large for a reliable determination of mean radial distances, coordination number, lattice, and vibrational disorder.

A tool to directly visualize contributions from different atomic species is wavelet transformation.^{117,118} This analysis utilizes the dependence of the backscattering amplitude on the photoelectron wavenumber and chemical element. As a rule of thumb, with increasing atomic number of the backscattering atom, the maximum backscattering amplitude occurs at higher wavenumbers. In Fig. 7(d), the wavelet transform (WT) of k^2 -weighted EXAFS measured at the Fe K absorption edge of a Fe-hosting polyoxopalladate sample is shown (spectrum presented in Fig. 7(c)). The WT clearly shows two maxima, around $k \approx 4 \text{ \AA}^{-1}$ and around $k \approx 9 \text{ \AA}^{-1}$. The first corresponds to oxygen, the latter to palladium atoms. Without any simulation or further data treatment, one can conclude that the mean pseudo-radial distance between Fe and oxygen is $(R + \Delta R)_{\text{Fe-O}} \approx 1.6 \text{ \AA}$ and between Fe and palladium $(R + \Delta R)_{\text{Fe-Pd}} \approx 3 \text{ \AA}$. This gives the possibility to monitor (time-dependent) structural changes around the same absorber for different elements separately.

In the last years, several programs and software solutions for WT in particular of EXAFS data have been released indicating the promising potential of this method. In the future, optimizing the wavelet for optimum resolution in k or real space, identification of the backscattering element and monitoring changes in the WT could benefit from machine learning algorithms.

Abbreviations

AFM	atomic force microscopy
AI	artificial intelligence
ALD	atomic layer deposition
APDMES	(3-aminopropyl)-dimethylethoxysilane
ASD	area selective deposition
ATP	adenosine-tri-phosphate
CMOS	complementary metal-oxide-semiconductor
CSD	chemical solution deposition
CNT	carbon nanotube
CNT-FET	carbon-nanotube field-effect transistor
CVD	chemical vapor deposition
DNA	deoxyribonucleic acid

EPE	edge-placement error
EXAFS	extended X-ray absorption fine structure
FT	Fourier transform
IC	integrated circuits
ICT	information and communications technology
IoE	internet of everything
IoT	internet of things
LCST	lower critical solution temperature
MEMS	microelectromechanical systems
MOD	metal-organic decomposition
NBC	network-based biocomputing
PNIPAM	poly(N-isopropylacrylamide)
NP	nanoparticle
PCB	printed circuit board
POM	polyoxometalate
PVD	physical vapor deposition
QD	quantum dot
RDF	radial distribution function
SAM	self-assembled monolayer
SMI	small molecule inhibitor
SSP	single source precursor
STM	scanning tunneling microscope
SWCNT	single-walled carbon nanotube
TRL	technology readiness level
TMDC	transition metal dichalcogenide
EUV	extreme ultra-violet
WT	wavelet transform
XANES	X-ray absorption near-edge structure
XAS	X-ray absorption spectroscopy
XMCD	X-ray magnetic circular dichroism
X(M)LD	X-ray (magnetic) linear dichroism

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