

Directed assembly of nanowires

Nanowires of a diverse range of compositions with tailored physical properties can be produced through synthetic means. These structures have been used as key components in flexible electronics, electronic logic gates, renewable energy technologies, and biological or gas sensing applications. Integrating these nanostructures into device or technology platforms will complement existing nanofabrication procedures by broadening the types of nanostructured materials that are utilized in device fabrication. This integration requires an ability to assemble these nanowires as controllable building blocks. Techniques are being developed that can quickly manipulate large quantities of nanowires through parallel processes.

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Fabrication using wire-like nanostructures is sometimes pursued out of necessity. An example is the high density of features required for state-of-the-art circuitry that dictates the widths of many electronic connections^{1,2}. However, nanowires can also be pursued for their properties that can be exploited in the fabrication of new functional materials or devices. A diverse range of properties is accessible through the numerous types of nanowires that can be produced by synthetic efforts³. Integrating these nanowires as key components will require the development of appropriate methods to assemble these materials. These techniques complement many different approaches to nanofabrication. This review will discuss a few of the approaches being pursued for directing the assembly of nanowires into various device architectures.

Developments in the synthesis and assembly of nanoscale materials will not supplant other efforts being pursued in nanofabrication. In fact,

much of our understanding of nanoscale materials is enhanced by the *synergistic efforts* of both bottom-up (e.g., synthesis and assembly)^{4,5} and top-down⁶ methods of making nanostructures. Quantum dots and quantum wires can now be produced in large quantities, and their band structure precisely engineered for desired properties^{7,8}. Another widely studied property is the surface plasmon resonance of metallic nanostructures, whose plasmon bands can be tuned from the visible to the near-infrared regions of the electromagnetic spectrum^{9,10}. However, nanostructures produced by top-down techniques are positioned within materials or devices during fabrication, and those nanostructures made by synthetic routes require further manipulation for integration into a similar material or device.

Is it worthwhile to develop techniques for manipulating nanoscale materials produced through synthesis if similar structures can be produced in the desired location through top-down approaches?

In order to answer this question, we must take a closer look at the differences between these two approaches. Top-down techniques can define the composition, shape, size and crystallinity of a nanostructure, as well as the lateral position of this structure on top of another material¹¹. These structures are fabricated through layer-by-layer processes; repeating a process of one material deposited over a second material followed by the selective removal of unwanted portions of either material¹². The resulting structures are easily integrated within the architecture of a device. Through synthetic control one can fine tune in three dimensions the composition, shape, crystallinity, and size of a nanostructure¹³⁻¹⁶. Many of these structures can be produced by synthesis with less effort (e.g., fewer hours of labor), lower cost (e.g., less material consumption and without the need for expensive high vacuum equipment), and in larger quantities (often more than a milligram is produced per synthesis) than through top-down techniques. Through a diverse range of synthetic techniques a wide range of materials is accessible; nanostructures have been synthesized from almost all of the transition metals, noble metals, and semiconductors^{3, 17, 18}. To capitalize on these benefits, it is essential to learn how to integrate the synthetic products into desired technology platforms¹⁹.

It is possible to grow nanowires directly on the surface of some materials and, therefore, avoid the necessity for assembling these structures. A few nanowires can be constructed with the desired shape, dimensions, and composition directly within the desired device platform²⁰⁻²⁴. These approaches are, however, limited to a few types of materials that can be assembled into a wire-like morphology. Isolating the synthesis of nanowires from their assembly into a device has many benefits, which include preserving the established reaction conditions for nanowire growth and purification without compromising the other materials within the final device. Furthermore, the synthesized nanowires can be manipulated as individual structures and assembled into complex architectures.

This review concentrates on techniques that have been developed to direct the assembly of many individual nanowires over large areas for their incorporation into various technologies or materials. Our focus is limited to the post-synthetic assembly of anisotropic nanostructures with nanoscale diameters and a wire-like morphology. Directing the assembly of these structures into desired configurations presents a unique challenge. The assembly of nanowires requires techniques that are able to appropriately manipulate and direct the deposition of individual nanostructures. Ideally these processes would also be cost effective and able to direct the assembly of many nanowires in parallel over large areas (several cm²).

Assembly by molecular forces

Many nanowires can be directly processed as a solution. Handling the nanowires as a solution is very convenient as these structures can be processed in a manner similar to most liquids. Also, the concentration of nanostructures can be adjusted through dilution. This suspension

of nanowires can be cast onto a substrate (Fig. 1). Nanowires that settle out of this solution will adopt random orientations. The strength of the interactions between the nanowire and the substrate determines both the adhesion of the nanowire to this substrate and the final orientation that will be adopted by this structure upon removal of the solvent. Interactions between the nanowire and the substrate can be enhanced through van der Waals and hydrogen bonding interactions.

A number of examples could be exploited to demonstrate the assembly and attachment of nanowires to a substrate using both hydrogen bonding and van der Waals interactions. Two common techniques include the hybridization of deoxyribonucleic acid (DNA), or protein-protein interactions to form protein complexes²⁵⁻²⁸. In either example, complimentary structures are linked to the surfaces of the nanowire and the substrate, respectively. The nanowire is bound to the substrate through the complex that is formed when the two complementary surfaces are brought into proximity. For example, a single-stranded DNA (or ssDNA) can bind selectively to the complementary DNA (cDNA) strand. This form of biorecognition can direct the assembly of nanostructures (Fig. 1)²⁵⁻²⁷. A droplet containing a suspension of DNA-modified nanowires is cast onto the substrate, and after DNA hybridization the nanowires are immobilized with random orientations either on or between the Au electrodes (Fig. 1b). The authors used hybrid Au-Pd nanowires to demonstrate the assembly of a hydrogen sensor²⁶.

Biorecognition is an efficient means for driving the assembly of nanoscale materials. It can be used to attach nanowires onto various surfaces, but this technique lacks the ability to direct the alignment of the nanowires during the assembly process. The alignment is driven largely by a shear force during solvent evaporation. The end application must be tolerant of misalignments. A further complication can be the reliance of this technique on the integrity of the biomolecules used in these examples. This process for directing the assembly of nanowires is, however, simple and can be easily adapted to many materials. Further modifications to this technique could utilize regioselective surface modification of nanowires using biotin-avidin linkages²⁸ or block-copolymer modifications²⁹, which have been pursued for end-to-end assembly of nanostructures. Another alternative is to take advantage of hydrophobic surface modifications that have been used to assemble composite nanowires into inverse micelle structures when immersed in aqueous environments³⁰.

Assembly by electrostatic interactions

Assembly of nanowires can be driven by electrostatic interactions³¹⁻³⁶. These interactions rely on either the inherent polarizability of the nanostructure or a surface modification of this material to adopt a specific charge. One example is the negative charge of silicon oxide at pH 7, that can attract nanowires with a positive charge (e.g., nanowires coated with an amine-terminated functionality)^{31, 32}. The negative

charge of silica can be partially masked by the selective patterning with a dielectric layer. An example of this modification is the printing of a methyl-terminated alkylsilane onto substrates of silicon oxide by micro-contact printing (μ -CP) as shown in Fig. 2a. Silicon nanowires functionalized with 3-aminopropyltriethoxysilane (APTES) adhere to the unmasked silica (Fig. 2b-c).

In the case that the nanowires have an inherent charge, these structures can be used without further modification to their surface chemistry. Some anisotropic nanostructures have an inherent charge due to the formation of a dipole within the material or residual charge from surface defects. Two such examples include divanadium pentoxide (V_2O_5) and zinc oxide (ZnO) nanowires. The V_2O_5 nanostructures are attracted to protonated surface of APTES³³⁻³⁶. For the ZnO structures the opposite behavior is observed; these

nanowires are attracted to a surface coating that is terminated with carboxylic acid functional groups or hydroxyl-terminated surfaces at high pH³². Interestingly, the ZnO nanowires were also shown to be attracted to surfaces receiving a negative electrical potential (e.g., -10 V). Large quantities of nanowires can be assembled onto the substrate with the assistance of an electric field. An electric field can also be used to assemble nanowires with surface modifications, such as the APTES coated silicon nanowires^{31, 32}.

Electrostatic interactions can assist the assembly of nanowires. This process can be driven by either passive or active (applied) electrostatic interactions, which are easily scalable over large areas. Simple structures are quickly assembled by these electrostatic interactions, but orientation of the nanowires can be easily altered by shear forces upon solvent evaporation.

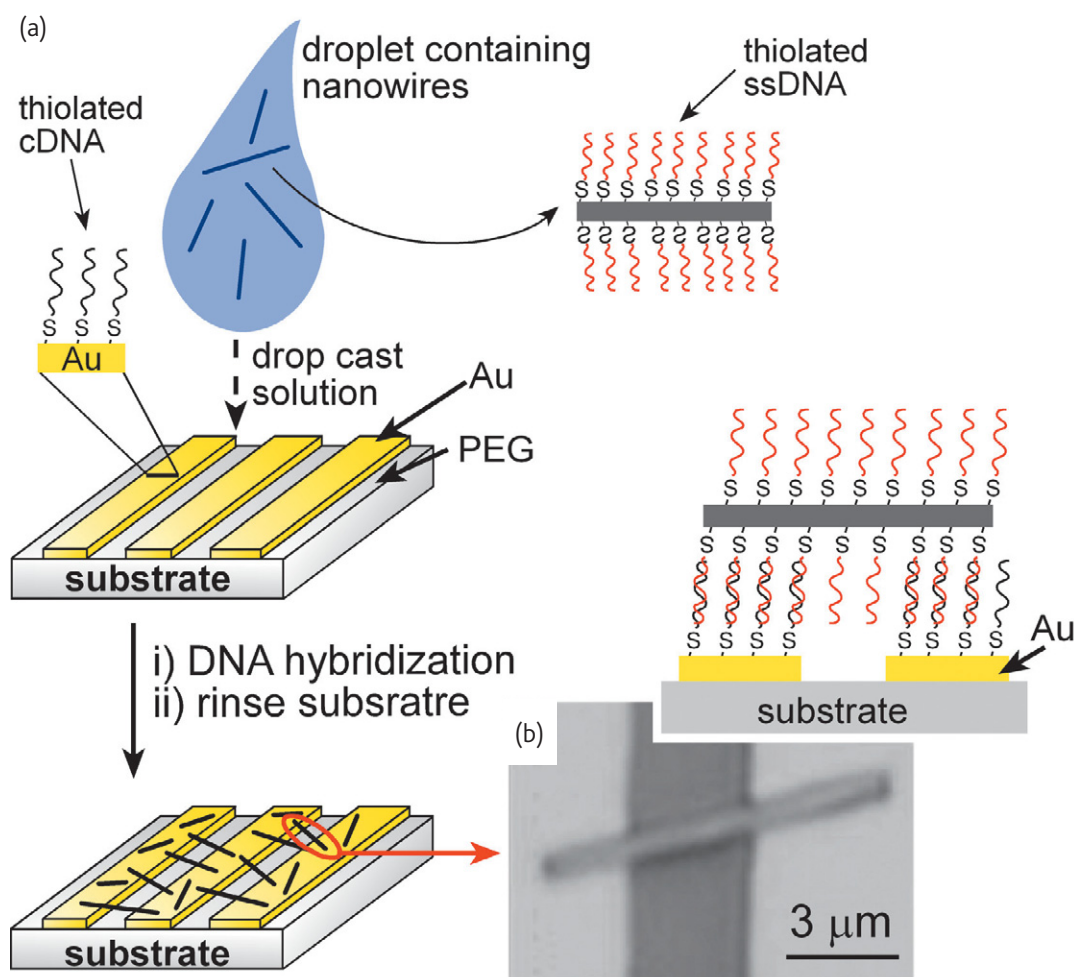


Fig. 1 (a) Assembly of nanowires can be directed by hybridization of DNA. A suspension of nanowires whose surface is modified with single-stranded DNA (ssDNA) is cast onto a surface patterned with the complementary DNA strands (cDNA) and polyethylene glycol (PEG). Nanowires bind to the substrate through hybridization of ssDNA with cDNA. After rinsing the substrate, nanowires adhering to the substrate can be imaged by (b) optical microscopy techniques. (Reprinted with permission from²⁶. © 2007 Wiley-VCH.)

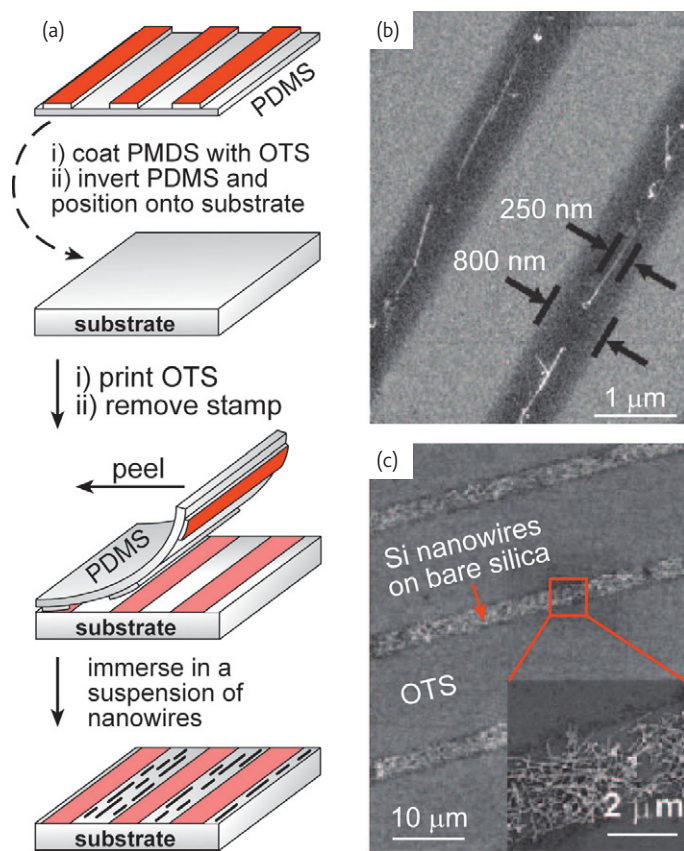


Fig. 2 (a) Nanowires can be assembled by electrostatic interactions. The negative charge of silica can attract ammonium-terminated nanowires. This negative charge can be selectively masked by patterning octadecyltrichlorosilane (OTS) by micro-contact printing (μ -CP) with a polydimethylsiloxane (PDMS) stamp. Scanning electron microscopy (SEM) images depict absorption (b) of individual nanowires and (c) bundles of nanowires onto exposed regions of silica. (Reprinted with permission from³⁷. © 2008 American Chemical Society.)

Assembly driven by shear forces

A shear force is created by the motion of a fluid or solid object against a second fluid or solid object. This motion can be achieved by a number of means, such as the wetting/dewetting of surfaces or the mechanical movement of two surfaces in opposite directions. In either case, a force is generated that is parallel to the direction of movement. This force can be used to redirect the orientation of the nanowires that are either suspended in a solution or fixed to a substrate. The new orientation of the nanowires will be parallel to the direction of the shear force.

The simplest demonstration of nanowire assembly assisted by a fluid flow is through the use of an evaporating liquid droplet (Fig. 3a). Nanowires deposited onto a substrate will reorient with the direction of the fluid flow to minimize the fluid drag³⁷⁻³⁹. Confining the fluid flow to a microfluidic channel can establish a shear force that is more uniform than that exhibited from an evaporating liquid droplet. This confined process has been used to align nanowires (Fig. 3b)⁴⁰⁻⁴³. These

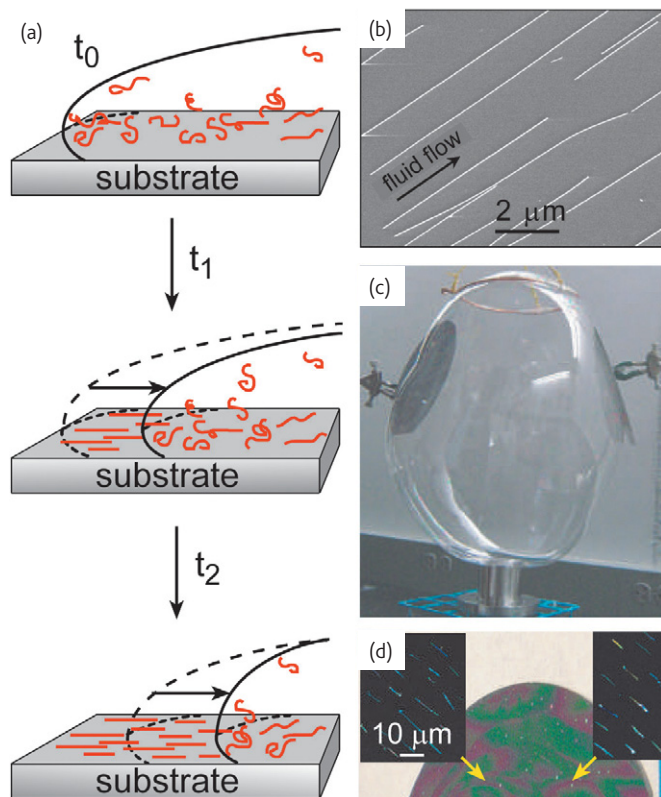


Fig. 3 (a) Shear forces from evaporation of a droplet can align nanowires with an orientation parallel to the fluid flow. (b) Fluid flow confined to microfluidic channels can direct the alignment, as shown in this SEM image of indium phosphide nanowires. (Reprinted with permission from⁴⁰ © AAAS.) (c, d) Photographs depict the assembly of nanowires over 100 mm wafers as the film of a bubble is drawn across the substrate. (Adapted from⁴⁴. © 2007 Nature Publishing Group.)

nanowires adhere to the surface and adopt an orientation parallel to the fluid flow. The density of nanowires can be controlled by changing the concentration of nanowires within the suspended liquid and flow time. Repeating this process after realigning the microfluidic channels can be used to create layer-by-layer assemblies of nanowires with complex geometries^{40, 41}. Subsequent steps of nanowire deposition require careful attention to the alignment of the microfluidic channel, as well as flow rates within the channel that can reorient nanowires deposited during the previous stages of assembly.

Applying a shear force over large areas requires a different approach. Nanowire assembly over length scales of an entire wafer can be achieved by passing the film of an air bubble across the wafer (Fig. 3c-d)^{44, 45}. This technique is inexpensive and can be adapted to patterning nanowires on many different types of substrates (e.g., flexible, flat or curved). However, a challenge for this technique is to control the viscosity of the bubble film in balance with a compatible surface coating on the nanowires, which has so far limited this demonstration to films of epoxy. Another challenge for this technique and for the alignment

achieved using microfluidic flow is to precisely control the position of the deposited nanowires.

The density of nanowires can be improved by adapting Langmuir-Blodgett (LB) techniques to align the nanowires⁴⁶⁻⁴⁹. Nanostructures suspended at an air-liquid interface can be assembled into dense films through compression within an LB trough (Fig. 4a). The nanowires adopt a close-packed organization to minimize the energy of the system during compression. Dip coating is used to transfer the nanostructures from the air-liquid interface onto a solid substrate. The wires can adhere to this substrate through van der Waals, hydrophobic-hydrophilic, or

electrostatic interactions as it is drawn in a vertical direction through the layer of compressed nanowires. A shear force is created along the vertical direction as the solvent withdraws from the surface of the substrate, leading to alignment of the nanowires along the same direction (Fig. 4b). The density of the deposited nanowires is controlled by the speed of withdrawing the substrate from the LB trough and the pressure applied to the assembly within the LB trough. Limitations of this technique include reorganization of the nanowires during dip coating that leads to overlapping features and gaps within the dense arrays of nanowires⁴⁹.

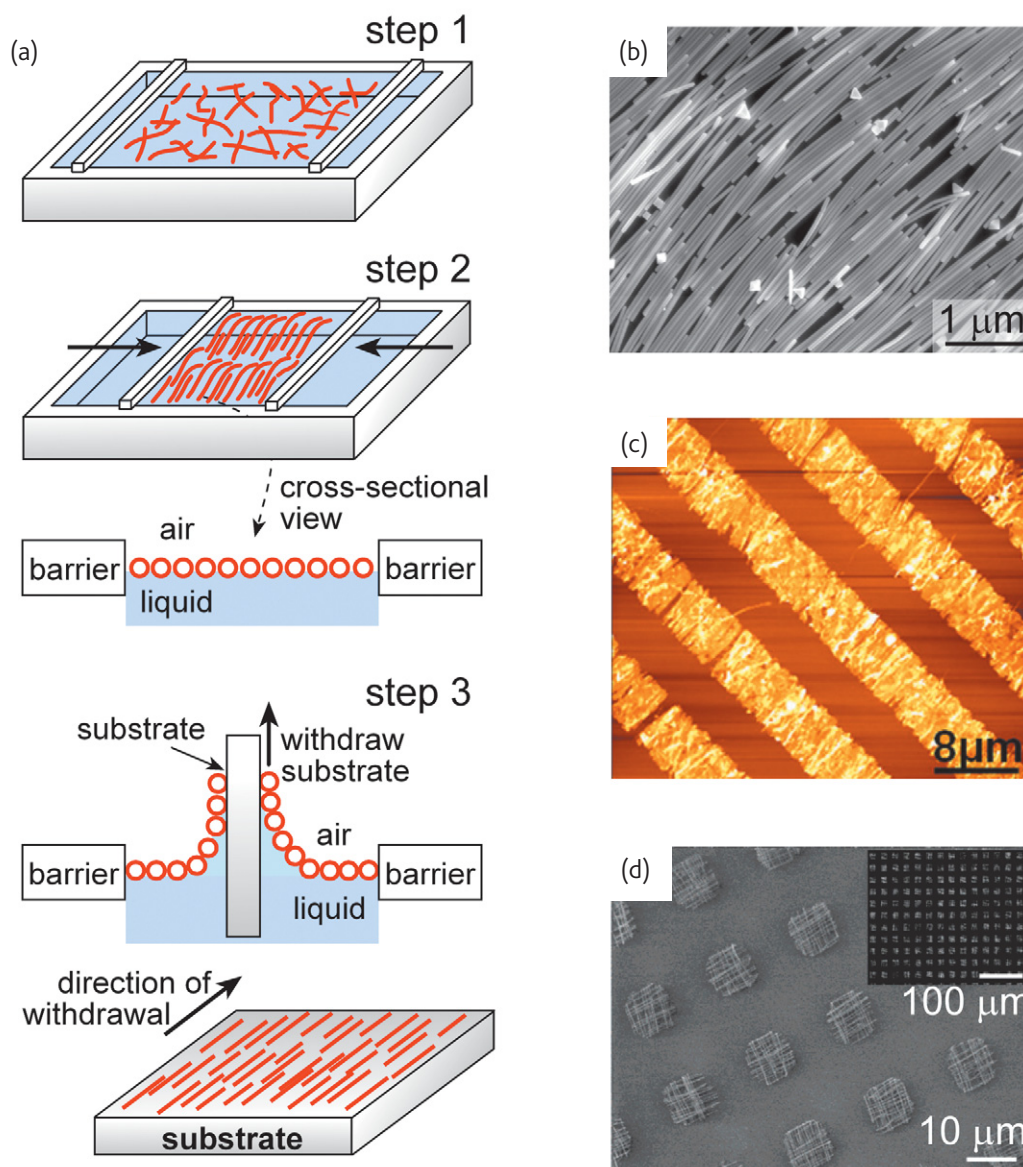


Fig. 4 (a) Nanowires can be assembled at an air-liquid interface within a Langmuir-Blodgett (LB) trough and transferred by dip-coating. (b) This SEM image shows an LB assembly of nanowires. (Reprinted with permission from⁴⁶. © 2003 American Chemical Society.) (c) The Langmuir-Schaefer (LS) technique can also assemble nanowires, as shown in this atomic force micrograph. (Reprinted with permission from⁴⁹. © 2008 Institute of Physics.) (d) The LS technique can assemble crossed arrays of nanowires, as shown in these SEM images. (Reprinted with permission from⁵¹. © 2003 American Chemical Society.)

Alternatively, the Langmuir-Schaefer (LS) technique can transfer uniformly dense arrays of nanowires from an air-liquid interface to the surface of a solid substrate. The LS technique positions the substrate so that its surface is parallel to the air-liquid interface. Horizontal transfer of nanowires onto the substrate prevents the reorientation of nanowires previously observed in dip coating using the LB technique. Alignment of nanowires is predominantly the result of isothermal compression of the wires at the air-liquid interface, and adhesion of the wires to the substrate is dominated by van der Waals interactions. The result of these effects is the transfer of close-packed arrays of nanowires that are more uniformly distributed across the receiving substrate than observed by the LB assembly process⁴⁹. These LS assembled nanowires have been incorporated as components within flexible electronics such as field-effect transistors (FETs)⁵⁰. This assembly process can be extended to substrates with a non-planar surface. For example, a polydimethylsiloxane (PDMS) stamp containing an array of parallel ridges and valleys, each with a rectangular cross-section, will transfer only the nanowires in contact with the raised ridges as this substrate is brought into contact with the air-liquid interface. These nanowires can be subsequently printed onto another substrate (Fig. 4c)⁴⁹. Hierarchical structures can also be produced by repeating the assembly process after changing the orientation of the substrate onto which the nanowires are being transferred. Crossed nanowire arrays are obtained by removing the unwanted regions of nanowires in preparation for integration within the device architectures (Fig. 4d)⁵¹.

The mechanical movement of two solid surfaces, one sliding across another, can also generate a shear force. This mechanically generated force can direct the assembly of vertically grown nanowires into arrays with an orientation parallel to the direction of the applied force (Fig. 5). A pattern can be imposed on the assembled arrays of nanowires by lift-off or similar lithographic patterning of these nanostructures (Fig. 5a)⁵². Nanowires with a crossed-wire configuration can be achieved by assembling the wires in two orthogonal directions (Fig. 5b-c)⁵³. This assembly technique can be scaled-up for high throughput. One approach is to create a shear force using a rolling substrate. Initial studies have demonstrated this process for continuous assembly of nanowires (Fig. 5d-e)⁵⁴. A common limitation for techniques relying on a shear force to assemble nanowires is the balance of the interacting forces. A minimum force is required to reorient the nanowires, and these structures must adhere to the substrate while maintaining their integrity.

Assembly within magnetic fields

A magnetic field can direct the alignment of magnetic nanowires suspended within a solution (Fig. 6a). Demonstrations have been limited to the assembly of nanowires composed of ferromagnetic and superparamagnetic materials⁵⁵⁻⁶⁰. The process of assembly can be guided by external magnetic fields, but the uniformity of the assembled patterns and control over positioning of the nanowires both

increase when magnets are incorporated onto the substrate. Nanowires assembled onto these substrates will often form chain-like structures extending from the magnet along its magnetic field lines (Fig. 6b)⁵⁵. Interestingly, a connected lattice of nanowires and micron-scale magnets can be assembled by a self-limiting process (Fig. 6c)⁵⁶. In this example, the nanowires span the distance between the fabricated bar magnets. Further demonstrations include the assembly of nanowires into crossed-wire configurations and assembly into hierarchical structures via magnets positioned beneath the substrate⁵⁹. A major limitation of this technique is the materials requirement that the nanowire being assembled must respond to a magnetic field. Future directions for this work could include the use of magnetic fluids to assist the assembly of non-magnetic nanowires.

Assembly by dielectrophoresis

Another area that has been explored for the parallel assembly of nanowires over large areas of a substrate is through the use of dielectrophoresis. Nanowires can adopt a polarization within an applied electric field. These structures align with a minimum energy configuration within the applied field, such as along the field lines (Fig. 7). This alignment depends on the field lines and their shape^{61, 62}. Theoretical understanding of how materials behave in an applied electric field is useful in designing systems for directing the assembly of nanowires⁶³. Nanowires have been assembled in high concentrations using dielectrophoresis (Fig. 7b)⁶⁴, or assembled as individual entities (Fig. 7c-d)⁶⁵. We have demonstrated the assembly of flexible, high aspect-ratio nanowires (Fig. 7e) over large distances (Fig. 7f-g)⁶⁶. These nanowires were assembled over distances > 1 cm. The assembly of nanowires by dielectrophoresis is an efficient process that can be applied to a number of materials (e.g., metals and semiconductors)^{61, 62, 64-73}. These structures can be assembled into functional devices, such as light emitting diodes^{67, 74} and resonators (Fig. 8a)⁶⁸. The process can be implemented over multiple length scales, but the remaining hindrance is the necessity of incorporating electrodes to direct the assembly process. Suspended nanowires also tend to bundle together either during or after dielectrophoretic assembly (e.g., during solvent evaporation). A promising approach to avoid the entanglement of nanowires during assembly is to encapsulate these structures within a matrix that can be solidified after completion of the assembly process⁷³.

Fabrication of devices from assembled nanowires

There are many potential applications that would benefit from the directed assembly of nanowires. The potential impact of this technology is probably the largest in the area of flexible electronics. The flexible platform used for flexible electronics are typically organic polymers that are not always compatible with the process conditions of many nanofabrication techniques. Techniques are being developed for the transfer of inorganic nanostructures to these flexible materials⁷⁵⁻⁷⁷,

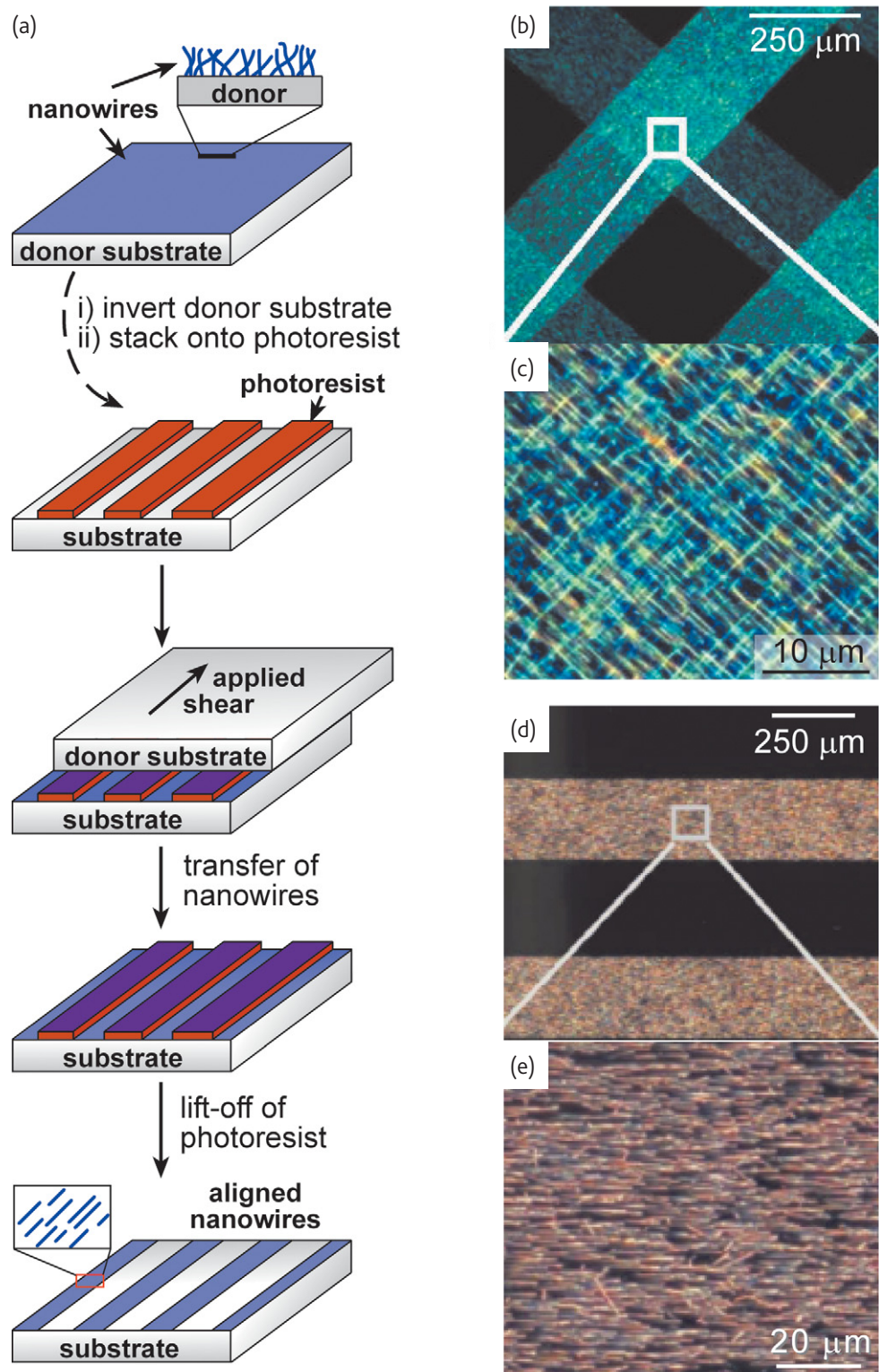


Fig. 5 (a) Mechanically applied shear forces will reorient nanowires along the direction of the applied force. Nanowires can be patterned by masking the substrate with photoresist. (b, c) Repeating this process in a perpendicular direction produces crossed silicon nanowires as shown in these dark-field optical microscopy images. (Reprinted with permission from⁵³. © 2008 American Chemical Society.) (d, e) Optical microscopy images show germanium nanowires assembled using shear forces applied through roll printing. (Reprinted with permission from⁵⁴. © 2007 American Institute of Physics.)

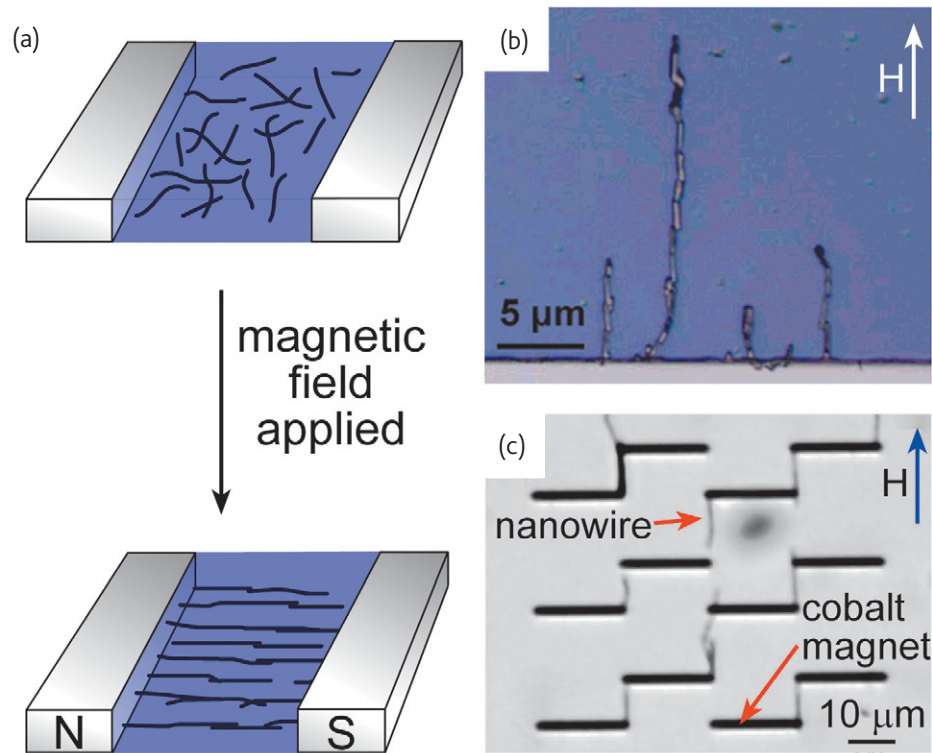


Fig. 6 (a) Superparamagnetic nanowires can be aligned within a magnetic field. (b) Composite Ni/Au/Ni or Ni/Bi/Ni nanowires self-align along magnetic field lines as shown in this optical microscopy image. (Reprinted with permission from⁵⁵. © 2005 American Chemical Society.) (c) Individual nickel nanowires can be assembled in a self-limiting fashion using arrays of patterned magnets as shown in this optical image. (Reprinted with permission from⁵⁶. © 2007 American Institute of Physics.)

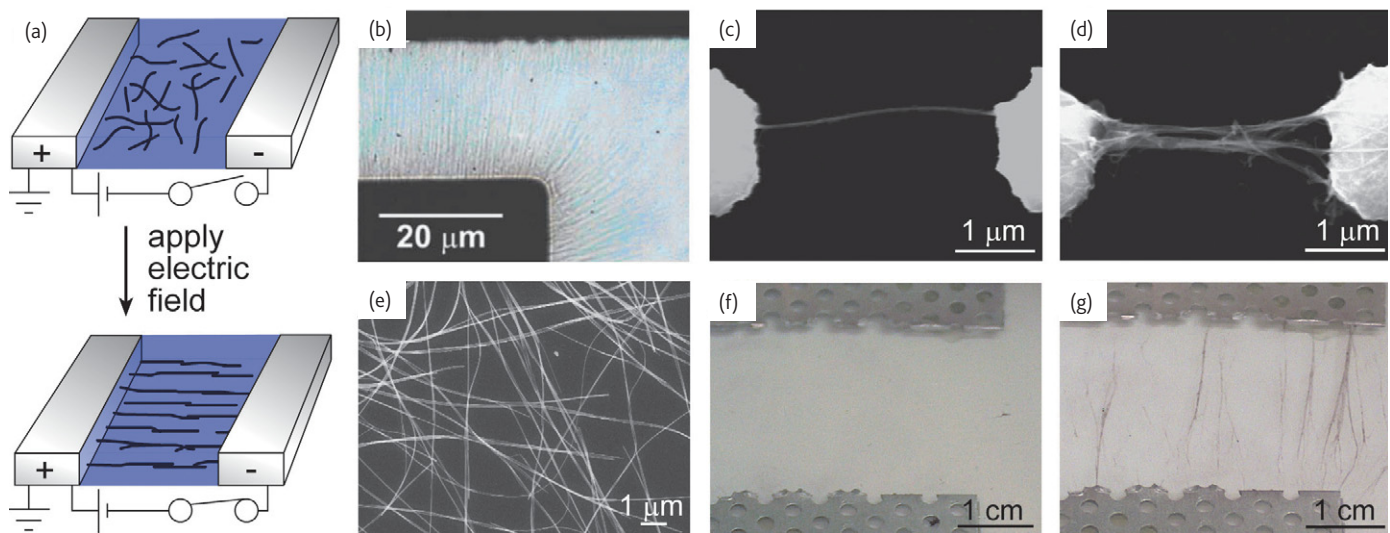


Fig. 7 (a) Polarization of nanowires in an electric field can direct the alignment of these structures. (b) Large quantities of cadmium selenide nanowires can be assembled as depicted in this optical image. (Reprinted with permission from⁶⁴. © 2007 American Institute of Physics.) (c, d) SEM images of zinc oxide nanowires assembled between electrodes. (Reprinted with permission from⁶⁵. © 2007 American Institute of Physics.) (e-g) SEM and optical images of flexible selenium nanowires (e, f) before and (g) after assembly.

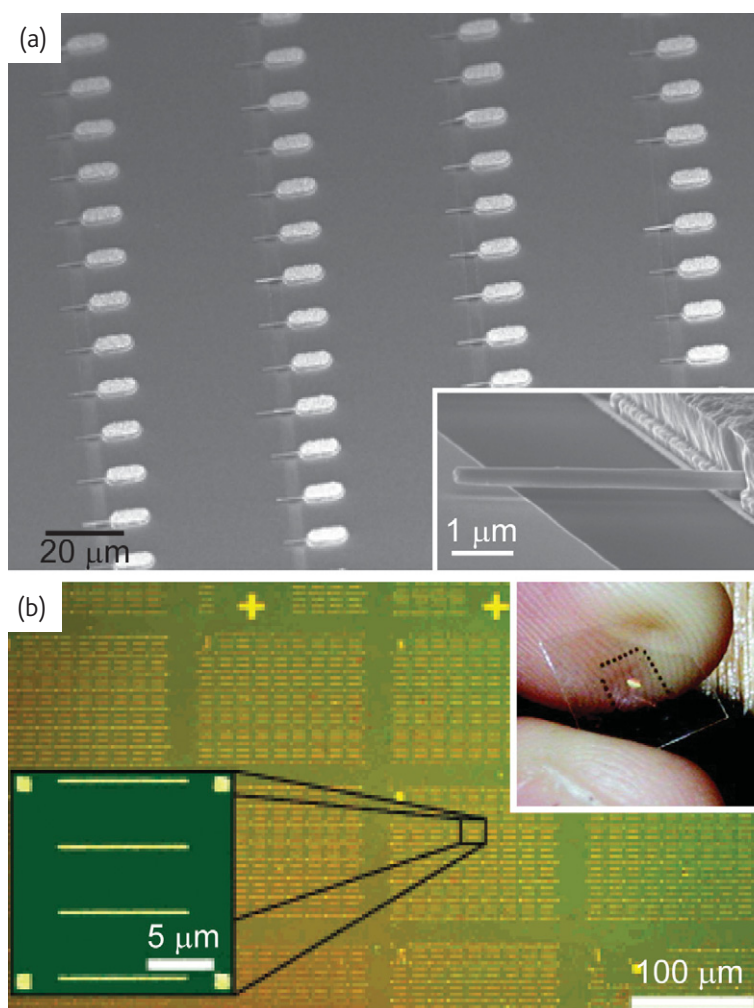


Fig. 8 (a) These SEM images show freestanding, resonant cantilevers fabricated from the assembly of nanowires by dielectrophoresis. (Reprinted with permission from⁶⁸. © 2008 Nature Publishing Group.) (b) Nanowires can be incorporated into flexible electronics. The deposition and alignment of silicon nanowires within these flexible architectures was directed by fluid-based shear forces. (Reprinted with permission from⁴². © 2003 American Chemical Society.)

as well as to establish robust electrical connections to these and other structures⁷⁸. Flexible electronics could benefit from the diversity of materials and properties offered through the synthesis and assembly of nanowires. Large-scale assemblies of nanowires directed by techniques outlined in this review are being used to fabricate devices or technology platforms through the incorporation of the wires as functional components. One of these demonstrations is the fabrication of nanoscale resonators (Fig. 8a) from the dielectrophoretic assembly of nanowires⁶⁸. Shear forces have also been used to direct the assembly of nanowires incorporated into the architecture of flexible devices (Fig. 8b)⁴². The utilization of nanowires in functioning devices is addressed elsewhere⁴.

Summary and outlook

Nanowires can be synthesized from a diverse range of functional materials, many with properties that can be either uniquely or easily achieved through synthetic processes. The assembly of these nanowires


can complement other nanofabrication techniques. In order to integrate these nanostructures into device or technology platforms, methods are required to direct the assembly of the nanowires. Suitable techniques should direct the assembly of nanowires over large area and manipulate large quantities of nanowires in a parallel process.

Nanowires have been successfully assembled as functional components in device or technology platforms. There are, however, several challenges that remain for efficiently directing the assembly of nanowires. These challenges include the manipulation of individual nanowires within a solution containing a high concentration of wires. In addition, each technique has a limited ability for lateral registration of the assembled nanowires. However, these techniques can be engineered to control the assembly of individual nanowires in parallel if the limitations of each technique are well understood. One example are resonators assembled by dielectrophoresis (Fig. 8a)⁶⁸. Upon assembling each nanowire the local electric field lines are

modified sufficiently to reduce the ability to manipulate other nanowires within the solution.

The assembly of high-aspect ratio nanowires presents another challenge. A material will behave more like 'cooked spaghetti' as the aspect-ratio increases. This flexible material does not respond uniformly to an applied force. Unraveling this material may require fixing one end of the nanowire while applying a shear force across the entangled structure.

Other challenges will be to achieve complex structures from assembled nanowires and assemblies with fewer defects. An efficient means of assembling nanowires in parallel could be pursued by combining many of the techniques presented herein. The appropriate combinations of techniques could create procedures with the ability

to efficiently assemble multiple wires and to also correct for defects. One approach could be large-scale assembly directed by shear forces, positioning of individual nanostructures using regioselective modification (e.g., DNA hybridization to one end of the nanowire), followed by self-alignment by dielectrophoresis or magnetism. 

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