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Nanofabrication using near-field optical probes

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Abstract

Nanofabrication using near-field optical probes is an established technique for rapid prototyping and automated maskless fabrication of nanostructured devices. In this review, we present the primary types of near-field probes and their physical processing mechanisms. Highlights of recent developments include improved resolution by optimizing the probe shape, incorporation of surface plasmonics in probe design, broader use in biological and magnetic storage applications, and increased throughput using probe arrays as well as high speed writing and patterning.

Keywords

nanofabrication; near-field scanning optical microscopy; NSOM; scanning probe; near-field imaging; lithography

Introduction

Nanofabrication is an essential component in many areas of current research, including semiconductor processing,¹ biomedical sensors and devices,² and photonic device engineering.³ Currently, ultraviolet photolithography is the mainstay of mass-produced nanofabricated devices and can yield half-pitch dimensions as small as 22 nm by combining various technologies such as 193 nm excimer laser exposure, immersion lithography, and double-patterning.¹ An alternative burgeoning technique for mass production is nanoimprint lithography,⁴ which has generated feature sizes as small as 3 nm. In both photo- and nanoimprint lithography, the fabrication of an expensive mask is required, which limits their use in prototyping and research. In these areas, the gold standard for maskless nanofabrication is electron beam lithography.⁵ However, it requires a large capital investment, and is designed to process materials in vacuum, which can make it incompatible with biological or gas and liquid phase materials.

Nanofabrication using near-field optical probes promises to be more cost-effective and versatile than electron beam lithography. Near-field probes are capable of confining light to arbitrarily small dimensions determined by the size of the probe, permitting them to surpass the conventional diffraction limit of approximately half the wavelength. These probes are widely used for imaging in near-field scanning optical microscopes (NSOMs / SNOMs),^{6–9} but have also been applied extensively for fabrication since the early 1990s.¹⁰ There are several different designs for near-field scanning probes, and here we review apertured tips, planar film apertures, apertureless tips, microspheres, and plasmonic lenses. In many cases,

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it is possible to parallelize these scanning probes to increase fabrication speed, and a few such studies will also be highlighted.

Both photophysical (thermal) and photochemical mechanisms have been harnessed to process materials. Through these broad classes of mechanisms, near-field optical probes can interact with the substrate and surrounding material by etching bulk surfaces, exposing photoresists, locally removing nanofilm coatings, and depositing molecules for the fabrication of both two- and three-dimensional devices. Many of these mechanisms are inaccessible via electron-beam lithography, making optical nanofabrication a highly versatile technique.

Near-field optical probe design

Canonical NSOM probes consist of a nanoscale aperture in a sharp tip either at the end of a tapered fiber, or through the sharp tip of a pyramidal atomic force microscope (AFM) probe (Figure 1a–c). Fiber-based tips are illuminated through direct coupling into the fiber, while pyramidal tips are usually free-space coupled from above with external optics, although a cantilever probe with an integrated light emitting diode and focusing lens has recently been fabricated.¹¹ In either case, an opaque thin metal coating is applied to the tip, which prevents optical transmission except through the aperture. Within the vicinity of the aperture, a strong evanescent field may be generated, with a small amount of light transmitted into the far field, where the transmission roughly scales as $(d/\lambda)^m$, where d is the aperture diameter, λ is the optical wavelength, and m is between 2 and 4 depending on the tip design.^{12,13} The light is confined with a lateral dimension roughly equal to the aperture size, and a longitudinal dimension that is generally of the order of a few tens of nanometers depending on both the wavelength and aperture diameter.

The aperture can be generated through deposition of the metal coating at a sharp angle, delicate mechanical abrasion of the tip, or for better controlled aperture shapes and sizes, via focused ion beam (FIB) milling. Using FIB milling, Jin and Xu have fabricated well-defined square apertures as small as 30 nm, along with a variety of other shapes.¹⁴ Head-on FIB milling allows the fabrication of arbitrary aperture shapes, while milling from the side provides greater control of the aperture size and surface quality.

Because the transmission falls off rapidly for decreasing aperture size, significant effort has been recently devoted to boosting the transmission for high-resolution probes while still providing tight confinement by optimizing the aperture shape. Apertures that incorporate a small lateral gap between two metal edges can have both tight optical confinement and high transmittance mediated by plasmonic antenna modes. C-shaped apertures were first explored, however bowtie-shaped apertures such as shown in Figure 1c have been found to be one of the most attractive choices so far.^{14,15} Combined theoretical and experimental studies have demonstrated that bowtie apertures with outline dimensions 260×260 nm are capable of generating intensity full widths at half-maximum of ~ 150 nm and peak intensities orders of magnitude larger than that found from circular apertures with similar optical confinement.¹⁶ Smaller bowtie apertures with outer dimension 160 nm can demonstrate even tighter fabricated line widths of e.g., 59 nm.¹⁷

Optical confinement and transmission efficiency can be further increased by using arrays of apertures or by fabricating a superstructure around an aperture. Kinzel and Xu found that a closely-spaced array of bowtie apertures with 20% open area allows extraordinary optical transmission (85%) through the structure due to waveguide modes and surface plasmon polaritons.¹⁸ Adding grating structures around isolated bowtie apertures (similar to Figure 1d) increased their transmission as much as 6-fold, and near-field intensity enhancements as much as 15-fold due to diffractive and surface plasmon polariton modes.¹⁹ In another study,

plasmonic grating structures were found to improve optical confinement by as much as 40%.²⁰ Appropriately designed grating structures alone, known as plasmonic lenses, are capable of confining light beyond the diffraction limit and can be used as near-field probes.^{21,22}

Near-field fabrication is also possible using probes that do not have an aperture. Apertureless tips have a resolution determined by their sharpness. As shown in Figure 1e, laser light is used to obliquely illuminate a sharp dielectric or metallic tip, inducing large localized intensities at the tip apex. Such a geometry is capable of generating orders of magnitude enhancement in intensity relative to the incident beam, with the greatest enhancement occurring when a metallic tip is proximal to a metallic surface.²³ These apertureless tips suffer from a larger background signal compared to apertured NSOM tips, but are capable of producing tighter optical confinement and enabling high-resolution AFM imaging *in situ*.^{24,25}

Specially designed, grating-coupled apertureless tips have also been fabricated. These tips can be considered a hybrid of apertured and apertureless tips because light is channeled to the tip using a waveguide surrounded by a reflective coating, but instead of the light exiting the tip through an aperture, it is coupled to the substrate via a nanoscale metallic plasmonic transducer.²⁶ This design provides increased coupling efficiency compared to a standard apertured NSOM probe.

Despite their relatively large size, spherical microbeads with submicron diameters can also be used as near-field probes, where light concentration to spot sizes smaller than $\lambda/2$ is due to a combination of focusing and evanescent modes. Typically, a long and narrow region of significant intensity enhancement is formed, sometimes referred to as a photonic nanojet. Although the original application of such probes was through their uncontrolled deposition in random or self-assembled close-packed arrays, a method of manipulating individual microspheres for surface patterning using a Bessel beam optical trap was recently devised by McLeod and Arnold (Figure 1f).²⁷ This method automatically establishes a nanoscale gap between the probe and substrate that is resistant to perturbations from surface roughness²⁸ or the dynamics of the patterning process.²⁹

A number of strategies have been developed for increasing the throughput of scanning near-field optical systems, many of which center around parallel arrays of probes. The major engineering challenge in such devices is maintaining a consistent separation between each probe tip and the substrate. An array of 16 cantilever probes with pyramidal tips was fabricated on the same chip by Haq *et al* (Figure 2a).^{30,31} In their device, the probes were in a linear configuration with a pitch of 125 μm . Individual probe tips could be addressed in parallel using either a spatial light modulator or a digital micromirror device. Other array implementations include a small sliding contact mask with bowtie apertures,³² and a set of metal-coated apertured soft polymer tips used in a tapping-mode based contact-expose-lift-translate sequence.³³ Although simpler to position, contact masks provide no direct control over the spacing between the probes and the substrate. A fixed regular array of microspheres fabricated using a specially-designed support layer and fluidic process was used to write patterns with as many as 400 beads simultaneously. Positioning this array required an intricate capacitive feedback system to maintain the parallelism and separation distance between the probes and the substrate.³⁴ Patterning using optically trapped microspheres has also been implemented in a limited array configuration, where each microsphere was trapped by its own Bessel beam, and could be used to write out patterns in parallel (Figure 2b).³⁵

An alternative method to boost throughput is to increase the writing speed of a single tip. One approach is to use light sliding contact, which eliminates the speed restrictions imposed by active feedback loops, and can therefore write at speeds of up to 10 mm/s.³⁶ At even higher speeds, Srituravanich *et al.* used an array of plasmonic hyperlenses on a flying head mounted above a disk spinning at 1000 rpm (Figure 2c).²² The head was aerodynamically designed to maintain a 20 nm separation from the disk surface, which was travelling at a relative speed of 4 to 12 m/s. They were able to write tracks on the disk surface as small as 80 nm. More recently, this technique has been improved by modifying the plasmonic lenses to include a C-shaped aperture at their center, which acts as an antenna to boost intensity and enhance optical confinement.³⁷ This effect, combined with advances in resist technology and the use of picosecond laser pulses allowed them to reduce the feature size to as small as 22 nm using a wavelength of 355 nm.

Material modification and fabrication mechanisms

Perhaps the simplest method of fabrication is via photophysical or photothermal interactions. In this mode, light is absorbed by the material surface, generating heat that leads to localized melting or ablation of the surface. Such processes are single-step, as they do not require etching or development to produce patterns, and have demonstrated some of the smallest feature sizes in near-field optical patterning. Chimmalgi, Grigoropoulos, and Komvopoulos wrote features with dimensions as small as 10–12 nm in thin metallic films using a femtosecond 800 nm laser and an apertureless NSOM system (Figure 3a).²⁴ Localized heating can produce either trenches or mounds,^{24,38,39} and can induce recrystallization of amorphous or polycrystalline materials, a technologically important process in the fabrication of nanoscale transistors.^{34,40} At higher laser pulse energies, ablation occurs, and spot sizes as small as 100 nm have been fabricated using 300 nm-diameter apertured tips and 400 nm femtosecond illumination.⁴¹ Nanoscale ablation and melting has also been demonstrated using a microsphere as the near-field probe, creating features on the order of 100 nm using 355 nm radiation.^{27,35}

Photothermal interactions can also be used to selectively deposit materials. Pan *et al.* demonstrated metallic nanoparticle sintering using a 532 nm laser and near-field microsphere-arrays to fabricate 100 nm feature sizes that could be reduced to 50 nm with additional post-processing (Figure 3b).³⁴ Such a technique is useful for fabricating nanoscale conductive traces, plasmonic devices, or surface-enhanced Raman scattering (SERS) devices.

Heat-assisted magnetic recording is a particularly promising application for photothermal material modification. For high density data storage, materials with a high coercivity, or resistance to demagnetization, are required to maintain a difference in magnetization between adjacent bits. However, high coercivity materials require a strong field for magnetization, which is difficult to generate at a small size scale. One potential solution is to use a laser to locally heat the magnetic material and temporarily lower its coercivity while the bit is being switched. A flying near-field optical probe has been used to generate this heating field using an 830 nm laser, enabling data to be written on a FePt nanofilm with a 70 nm track width.²⁶ Further theoretical studies predict the possibility of reducing the magnetic bit size to 20 nm using optimized bowtie-shaped probes.¹⁵

Although not the simplest, the most widely used near-field nanofabrication mechanism is the exposure of a photoresist thin film. Exposure to light causes a chemical change that, upon development, causes selective removal (positive resists) or adhesion (negative resists) of the photoresist film. In general, patterning the photoresist is not the end goal as it was for the photophysical techniques above, but is a single step in the patterning or deposition of other

materials. Photoresist-based processing formed the first demonstration of the use of NSOM probes for nanofabrication, where line widths of order 100 nm were demonstrated using 420 nm light.¹⁰

Ultra-thin resists provide the best resolution for near-field techniques because the entire resist thickness remains within the rapidly diverging optical near-field. To this end, a significant amount of work has gone into developing self-assembled monolayer (SAM) photoresists. As depicted in Figure 3c, SAMs typically consist of short-chain organic polymers with a head group (often sulfur) that has a strong affinity for the substrate (a noble metal, often gold). Mercury-based and silane chemistries represent other potential SAMs for nanopatterning. As these films consist of only a single monolayer, they can be limited to nanometer-scale thicknesses. The first demonstration of near-field patterning of a SAM was in 2002 where lines as small as 40 nm were repeatably written by UV-induced oxidation of a functional group on the SAM molecule.^{30,42}

More recently, near field patterning of SAMs has focused on reducing the feature size to as small as 9 nm,⁴³ and using the technique to fabricate devices out of a variety of biomolecules. Here, SAM molecules are chemically activated by exposure to light, enabling binding with the target biomolecule, as shown in Figure 3c. Patterns of tethered DNA molecules with linewidth under 100 nm have been created in this way,^{44,45} as well as lines of yellow fluorescent protein with a width of ~200 nm,⁴⁶ lines of immunoglobulin G protein molecules as narrow as 80 nm,⁴³ and optically active photosynthetic proteins with 98 nm resolution.⁴⁷ Patterned biomolecules can serve as sensors or as platforms to study communication and energy transfer between biomolecules.

Beyond the solid-state photochemical reactions typically harnessed in photoresist-based processing, optical near fields can be used to direct liquid and gas-phase chemical reactions. There have been a limited number of such studies recently, with the most immediate results being a decade old. A tapered apertureless fiber tip was used to concentrate 514.5 nm light and etch 30 nm wide patterns on silicon in a Cl₂ atmosphere.⁴⁸ Another example involves the deposition of Zn on a glass substrate from the precursor gas diethyl-zinc using an NSOM probe, where feature sizes of 20 nm were fabricated using a 244 nm laser wavelength to define nucleation sites.⁴⁹

Photochemical interactions form the foundation of photopolymerization, which has been used extensively to build three-dimensional structures out of a resin that hardens upon exposure to ultraviolet light.⁵⁰ In the conventional far-field implementation, nonlinear absorption of infrared light is used so that the polymerization is limited to individual voxels and does not occur along the entire beam path. In the near-field implementation, the polymerization occurs immediately at the probe tip, and therefore direct linear absorption can be used. In one example, a self-waveguiding optical tip with high aspect ratio was fabricated using pulses of 405 nm light at the end of an optical fiber.⁵¹

Summary

Near-field optical probes modify materials for nanofabrication with feature sizes as small as <10 nm—almost two orders of magnitude below the illumination wavelength. Various near-field probe designs have been reviewed here, along with a number of different photophysical and photochemical mechanisms. As there is not much further room for major improvements in resolution, future development is expected primarily on increasing the light throughput via larger arrays of particles and higher writing speeds, and on broadening the range of applications and types of materials that can be processed. Better demonstrations of true 3D fabrication and integration of this technique with micro- and meso-scale fabrication

techniques for hierarchical device assembly will also further increase the overall impact of near-field optical nanofabrication techniques.

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Figure 1. Near-field optical probes

(a) An aluminum-coated tapered optical fiber with 70 nm aperture at the tip apex. Reproduced from ¹⁰. (b) Optical micrograph of the tip in (a) showing 488 nm emission from the tip apex. Reproduced from ¹⁰. (c) Bowtie-shaped aperture carved in the apex of a pyramidal AFM tip using FIB. Reproduced from ¹⁴. (d) Plasmonic lens combined with a dumbbell-shaped aperture fabricated on a metallic thin film. Reproduced from ³⁷. (e) Schematic of an apertureless probe tip with oblique illumination. Reproduced from ²³. (f) Diagram of an optically trapped microsphere used as a near-field optical probe for substrate nanopatterning. Reproduced from ²⁹.

Figure 2. Increasing throughput

(a) An array of apertured NSOM tips mounted on cantilevers. Reproduced from ³⁰. (b) Patterns written simultaneously using an array of optically trapped microspheres. The different linewidth is due to different bead sizes. Scale bar is 2 μm. Reproduced from ³⁵. (c) A flying near-field optical probe used for high-speed nanofabrication. Reproduced from ²².

Figure 3. Fabrication mechanisms

(a) Nanoscale photothermal writing of lines in a 25 nm gold film using an apertureless near-field probe. Surface relief measured using AFM. Reproduced from ²⁴. (b) Schematic of using near-field probes to sinter or ablate gold nanoparticles depending on the laser energy. Reproduced from ³⁴. (c) Schematic of the photochemical processes involved in patterning SAMs and attaching protein molecules. Reproduced from ⁴⁷.