

Near-Field Scanning Optical Nanolithography with Surface-Wave Enhanced Probes

Reuben T. Collins, Cecile Veauvy, Ian C. Schick, P. David Flammer, Michael A. Hurowitz, James T. Martineau, Evan J. Schick; Colorado School of Mines; Golden, CO; Russell E. Hollingsworth, Gregory J. Nuebel; ITN Energy Systems, Inc.; Littleton, CO

Abstract

An optical approach to nano-patterning using near field scanning optical microscopy (NSOM) is presented. Using an NSOM specifically designed for patterning 4 inch wafers, we have demonstrated sub-100 nm feature sizes in conventional i-line photoresists, optical patterning of a-Si:H based resists, and patterning over large distances without stitching errors. A series of test structures were also fabricated including quantum point contacts and micro crystallite arrays. Novel NSOM probes based on excitation of surface plasmon polaritons are also being explored. The higher throughput and lower damage threshold possible with this approach suggests a significant increase in write speed over conventional NSOM probes can be achieved.

Introduction

Advanced lithography has been the key technology driver for the semiconductor industry with sub-100 nm feature sizes now in manufactured products. As dimensions shrink, new resists, device designs, processes for making interconnects, and techniques for doping and etching must be developed. Research grade lithography tools that allow new processes, materials, and devices to be tested for sub-100 nm performance are essential to each of these advances. Direct write lithography using scanning techniques is ideally suited to this application. E-beam lithography allows feature sizes down to several tens of nanometers and has been the most commonly used technique for research structures. On the other hand, many scanning probe techniques have been adapted as lithography tools and several of them, namely STM and AFM, have given very encouraging results (see for example Ref. [1] and references therein)

This paper discusses an alternative approach based on near-field scanning optical microscopy (NSOM).[2] NSOM is a scanning probe technique like STM or AFM. In the case of NSOM, the probe is an optical fiber which is either etched, milled, or thermally pulled to a point. The probe is coated with metal leaving a small, nanometer-sized aperture at the tip. In our microscope, UV light from an argon ion laser is coupled into the probe and used as a light source to expose an optical photoresist. Patterns are written by scanning a resist coated sample beneath the probe while maintaining a fixed tip-sample separation of less than an optical wavelength, in the near-field, using shear-force feedback.[3] Working in the near-field with a subwavelength aperture allows features much smaller than the diffraction limit to be patterned. Two instruments were used in this study. The first is a conventional metrological NSOM with a scan range of 100x100 μm^2 . The second was specifically designed for lithography, and the scanning stages use very high resolution closed loop control allowing 100 mm continuous travel with position resolution better than 20 nm.[4] In addition to a patterning tool, it

functions as a high resolution metrological tool allowing nanoscale topographic and optical (e.g. reflection and transmission) images of patterned regions to be acquired.

Relative to e-beam lithography, NSOM lithography represents a lower instrument cost without need for a vacuum system or low electrical noise facility. Near-field patterning can be done in a larger range of ambients including air and on non-conducting substrates. The approach also avoids radiation damage from high energy electrons. NSOM-based lithography may also have advantages for pattern registration. Multi-level patterning requires imaging of alignment marks. Imaging in e-beam lithography results in exposure of the resist that overlays the marks which can lead to blurring of the alignment marks. In an NSOM, imaging with red light allows resist covered features to be optically and topographically scanned, and patterns aligned, without unwanted exposure. It may also be easier to integrate conventional optical lithographic patterning and processing with NSOM patterning.

Two potential limitations of NSOM lithography are resolution and write speed. Spatial resolutions in optical measurements of less than 20 nm have been reported in NSOM,[5] however, 50 nm is a more realistic lower lithography limit using conventional tips. Write speed is limited by the low optical throughput of the subwavelength aperture and the damage threshold of the metal coating. Below we discuss a new approach to making tips based on excitation of surface plasmon polariton (SPP) modes which increase optical transmission and are less sensitive to damage. Increased transmission can have the secondary effect of allowing smaller apertures, hence, improving resolution.

NSOM Patterning and Performance

Metrology

In topographic imaging, the NSOM developed for lithography shows spatial resolutions comparable to commercial NSOMs. The large travel range of the instrument, however, is unique as demonstrated in Fig. 1a which shows a topographic image of a pattern of 80 nm tall gold lines on a silicon wafer. The dimension of the image is $1 \times 0.7 \text{ mm}^2$ and it was acquired as a single image without any stitching. As a comparison, a schematic rectangle defines the maximum scan range of most commercial scanning probe microscopes. Figure 1b presents simultaneous topographic and reflection images of a smaller region of the sample demonstrating the same contrast in both images.

Patterning with conventional photoresists

Several reports of NSOM based lithography have previously been made demonstrating features smaller than a few 100 nm in conventional[6] and unconventional[7] photoresists. Based on minimizing resist thickness and critical dimension and having off-the-shelf availability, we explored NSOM patterning using a

commercially available i-line photoresist, Shipley SPR505A, which was typically thinned 50% in ethyl lactate to give a thickness of ~ 150 nm after spinning. This thickness was chosen because it was thin enough to pattern very small features but thick enough to withstand most standard lithography post-process etches and to allow metal lift-off.

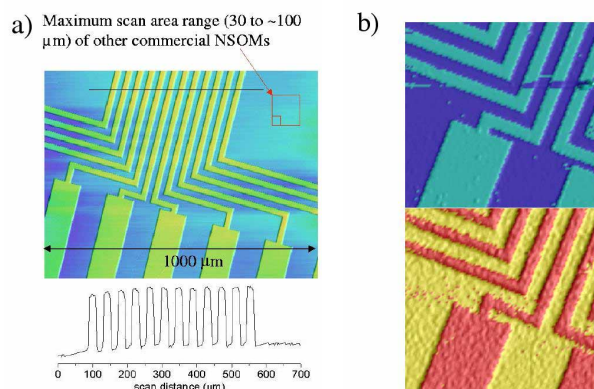


Figure 1 a) NSOM topographic image of a pattern of 80 nm tall gold lines on silicon. The image size is 0.7×1 mm² with 2 µm point spacing. The horizontal line cut was taken from near the top of the image. b) Simultaneous topographic (upper) and reflection (lower) images of a 300×300 µm² area of the pattern.

As a test, sets of lines were patterned into the resist for write speeds from $0.5 \mu\text{m s}^{-1}$ to $3 \mu\text{m s}^{-1}$ using fixed exposure intensity. Figure 2a shows an AFM image of a set of lines for a writing speed of $2 \mu\text{m s}^{-1}$. Line widths and shape are homogeneous. The cross sectional profile shows a line width of 300 nm. Widths down to

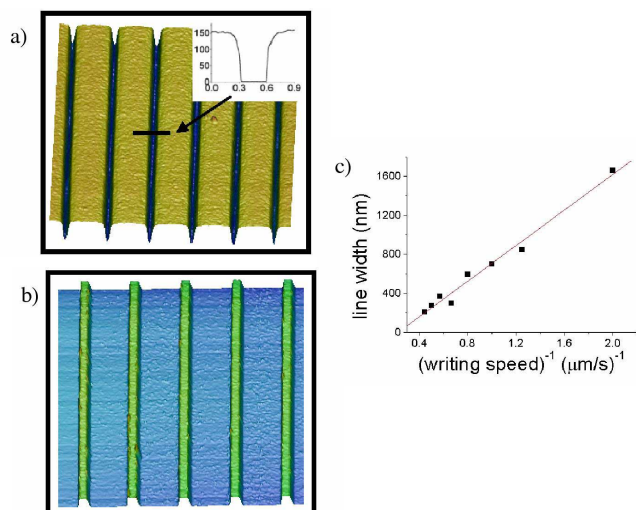


Figure 2 a) AFM image of ~ 300 nm wide lines written into Shipley SPR505A. The spacing between lines is 2 µm. b) AFM image of gold lines made by 30 nm thick gold lift off on the previous sample. c) The line width of gold lines following lift-off as a function of the inverse scan speed (equivalent to the optical dose).

200 nm with flat bottoms and the sharp sidewalls were routinely produced. When lines were prepared with widths below 100nm, however, the edges appeared rough. High resolution imaging of the photoresist layer itself after development indicated it had a texture of several tens of nanometers which was responsible for this roughness. Lift-off of a 30 nm gold film deposited on the sample in Fig. 2a led to the gold lines in Fig. 2b. Figure 2c shows the line width follows a linear dependence with inverse write speed, indicating that feature size is proportional to the optical dose. Lift off tended to allow smaller features with gaps between features of 70 nm successfully demonstrated.

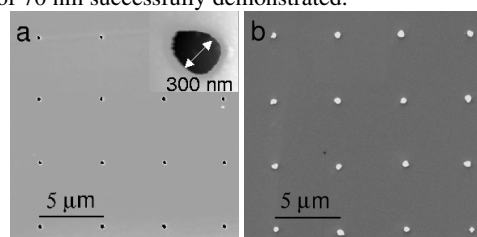


Figure 3 a) AFM image of a hole array patterned in photoresist. The insert shows a blowup of one of the holes. The shape is a function of aperture shape. b) SEM image of a GaAs dot array.

This patterning process was used in several applications including the creation of regular arrays of GaAs micro-crystallites[8] as shown in Fig. 3, and fabrication of gated quantum point contacts in a 2-dimensional electron gas which exhibited quantized conductance.[4] The latter was interesting because it combined conventional optical lithography with NSOM lithographic patterning in a single resist layer, and involved using NSOM topographic measurements to align the near-field and conventional patterns.

Long travel capability is illustrated in Fig.4 which shows an SEM micrograph of 1000 µm tall gold letters with 400 nm line widths prepared by gold lift off. Each line segment was written in a continuous scan with no stitching.

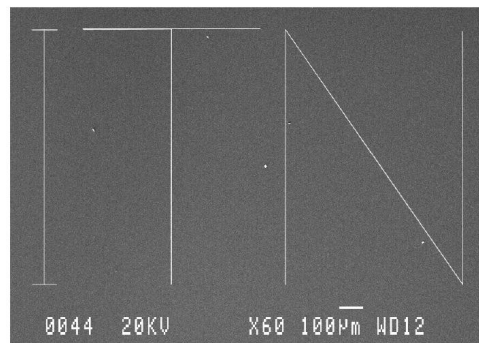


Figure 4 SEM image of one millimeter tall gold letters written by near-field lithography without stitching using a liftoff process and SPR505A i-line resist.

Unconventional Photoresists

Amorphous silicon has been shown to be a potentially useful resist for nanoscale lithography based on scanning microscope techniques.[9] In general, patterning a-Si:H involves selectively

oxidizing the a-Si:H surface by local removal of hydrogen passivation. The oxide layer then becomes a mask for subsequent etching of the surface.

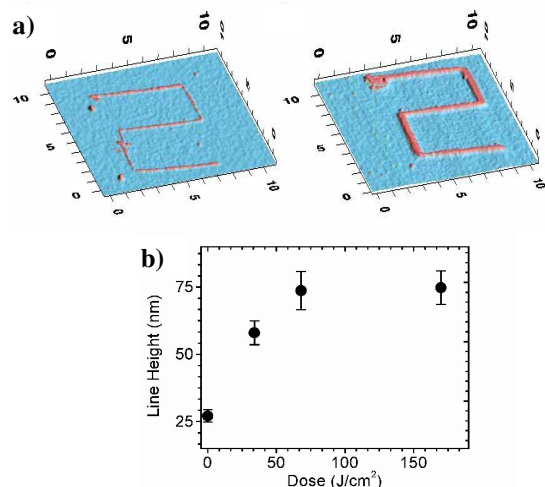


Figure 5 a) AFM images of test patterns generated by proximity of the NSOM tip to the a-Si:H surface in the absence of illumination. Dimensions are in microns. The tip is dithered as part of the shear force feedback used to maintain constant tip/sample separation. Increased dither amplitude in the right image relative to the left resulted in larger line width. b) The dependence of line height on optical exposure for fixed dither amplitude. Patterns are more fully developed as the optical exposure is increased.

In our work, optical illumination of the a-Si:H by the NSOM tip was used to selectively oxidize the surface, although it was also found that proximity of the tip to the surface in the absence of light resulted in partial oxidation of the layer and pattern formation.[10] A hydrogen plasma etch was used to *develop* patterns written into the a-Si:H resist. Fig. 5a shows test patterns written using proximity patterning without light. Line widths of ~100nm, comparable to the tip size, were written in this way. The height of the lines after development, however, was less than the original a-Si:H layer thickness, indicating incomplete exposure (oxidation). In the presence of illumination, the line heights increased as shown in Fig. 5b. The doses required for pattern heights to reach the full film thickness were consistent with doses determined from far field optical studies of the a-Si:H resist.

Plasmon Enhanced NSOM probes

One of the central issues in NSOM lithography is write speed which in our studies is limited by the optical dose that can be delivered by the tip. The transmission coefficient of a small diameter hole in a thin metal film is predicted to follow a $(d/\lambda)^4$ dependence where d is aperture diameter and λ is wavelength.[11] For the sub-100 nm apertures of metallized NSOM fiber probes, actual transmission efficiencies are typically in the 10^{-6} or less range, although there are reports of efficiencies approaching 10^{-3} for tips with an optimized taper.[12] Transmission by itself would not be an issue if input power had no limitation. One side effect of the process of tapering a fiber, however, is reduction of the diameter of the fiber core resulting in loss of guiding with most of the power leaking out of the fiber. The metal coating applied to fiber is intended to block this light and insure transmission comes

from only the aperture. The leakage has the effect of focusing nearly all of the input power on a small region of the metallization near the fiber point where it heats the metal. This empirically limits maximum input power to ~1mW to avoid thermal damage.[13] With a transmission efficiency of 10^{-4} and a required dose of 50mJ/cm² (consistent with SPR505A), a 100 nm aperture would have a maximum write speed of 2mm/s. This is adequate for many basic research structures, but far too slow to pattern a significant fraction of a wafer. At higher resolutions we expect the transmission to drop off rapidly from the $(d/\lambda)^4$ dependence further suppressing write speed.

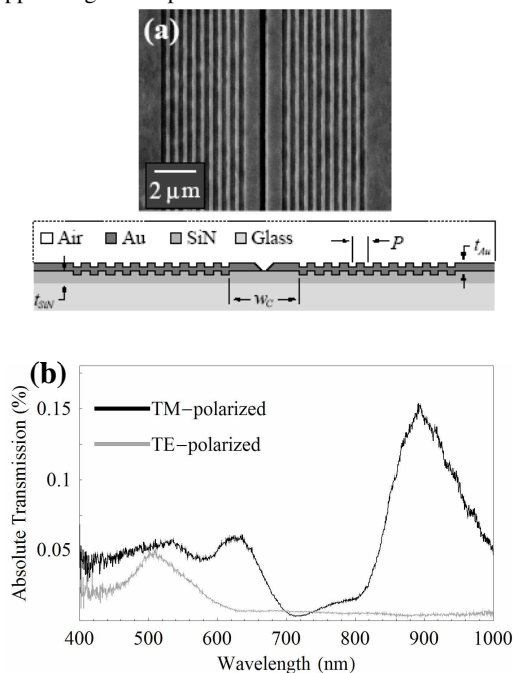


Figure 6 a) SEM image and schematic of a linear aperture in a Au film flanked by periodic arrays of grooves. b) Polarized transmission spectra for a structure of the type shown in a) with parameters $t_{Au}=200$ nm, $t_{SiN}=200$ nm, $P=400$ nm, $w_G=1042$ nm. There were 10 grooves, 75 nm deep on each side. Aperture width was 80 nm on the SiN side and 450 nm on the air side.

Recent studies of subwavelength apertures in metal films which are surrounded by periodic grating structures have demonstrated transmission enhancements of roughly an order of magnitude relative to an isolated aperture.[14] An example of such a structure, shown in the SEM image and schematic in Fig. 6a, involves a linear aperture flanked on each side by a periodic array of grooves. This structure was patterned using a combination of e-beam lithography and broad area ion milling. Fig. 6b shows transmission spectra for such a structure with the electric field polarized parallel (TE) and perpendicular (TM) to the grooves. In TM-polarization, a strong transmission suppression is visible from 700-800 nm with a strong enhancement near 900nm. The wavelengths of enhancement and suppression can be tuned by changing the separation between the groove array and the aperture. This effect arises when surface plasmon polariton (SPP) modes generated by the grating structure interfere with the incident light creating regions of high and low power flow at the metal film surface.[15] At wavelengths where maximum power flow occurs

at the location of the aperture, transmission is enhanced. Resonant cavity effects from reflection of the SPP modes off of the grating structures further contribute to this enhancement.

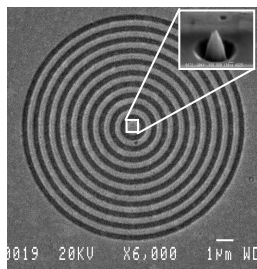


Figure 7 A fabricated bulls-eye grating and post feedback extension for a plasmon enhanced NSOM probe.

Figure 7 shows a bulls-eye grating structure analog of the linear array in Fig. 6 which might be suitable for fabrication on the cleaved end of an optical fiber. With this design, optimizing the SPP interference and cavity effects may allow transmission enhancements much larger than an order of magnitude to be realized. Equally significant, optical damage thresholds should be much higher than for conventional probes without the concentrating effect of the tip taper. The structure in Fig. 7 illustrates incorporation of a metal point to allow shear force feedback regulation of the separation between the probe and the sample. Electromagnetic field modeling has been used extensively to assist in understanding and optimizing these structures. Figure 8, for example, shows power flow and intensity for a 2-d structure with a metal tip, demonstrating that the fields actually concentrate around the tip.

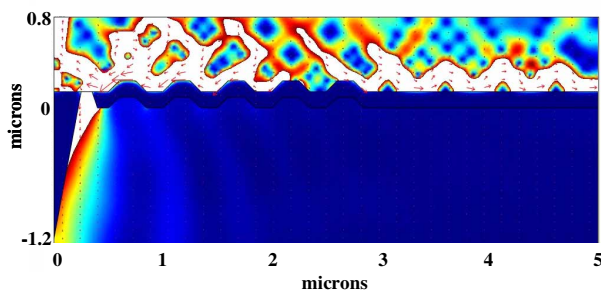


Figure 7 Numerical simulation of the power flow intensity (false color scale with blue as minimum and white as maximum) and direction (arrows) for a plane wave illuminating a 2-d grating structure in a gold film on glass. Mirror symmetry allows computation to be simplified by modeling half of the structure. This is what is shown. A metal post extension (left side) and aperture next to the metal post have been included. The grating period is 500nm. Illumination is from the glass side (top) at $\lambda=800$ nm which is the wavelength of resonant transmission. The transmitted power is heavily concentrated around the gold post surface.

Conclusions and Acknowledgements

We have demonstrated that near field microscopy can be used to fabricate sub-100 nm features in both conventional optical photoresists and unconventional a-Si:H resists and have shown that SPP enhanced probes may enable high throughput optical probes.

Not only does NSOM lithography extend the useful range for optical resists, it also avoids the radiation damage and expense of e-beam lithography while allowing for easy patterning of non-conducting substrates under ambient conditions.

The author's gratefully acknowledge valuable discussions with and assistance of Sheena Murphy, James Hicks, Matthew Johnson, Mary Herndon, Matthew Treaster, Tom Kuech, Joseph Beach, John Yarbrough and Cary Allen. This material is based upon work supported by the National Science Foundation under Grant Nos. DMI-0110486 and DMI-0522281.

References

- [1] A. A. Tseng, A. Notargiacomo, and T. P. Chen, J. Vac. Sci. Technol. B 23, 877 (2005).
- [2] E. Betzig, J.K. Trautman, T.D. Harris, J.S. Weiner and R.L. Kostelak, Science, 251, 1468 (1991).
- [3] K. Karrai and R.D. Grober, Ultramicroscopy 61, 197 (1995).
- [4] C. Veauvy, M.A. Treaster, R. T. Collins, R. E. Hollingsworth, J. L. Hicks, and S. Q. Murphy, submitted Appl. Phys. Lett.
- [5] E. Betzig and J. K. Trautman, Science 257, 189 (1992).
- [6] L.P. Ghislain, V.B. Elings, K.B. Crozier, S.R. Manalis, S.C. Minne, K. Wilder, G.S. Kino and C.F. Quate, Appl. Phys. Lett., 74, 501 (1999); I. I. Smolyaninov, D. L. Mazzoni, and C. C. Davis, Appl. Phys. Lett. 67, 3859 (1995); X. Yin, N. Fang, X. Zhang, I. B. Martini and B. J. Schwartz, Appl. Phys. Lett. 81, 3663 (2002).
- [7] S. Davy and M. Spajer, Appl. Phys. Lett. 69, 3306 (1996); R. Riehn, A. Charas, J. Morgado and F. Cacialli, Appl. Phys. Lett., 82, 526 (2003); S. Madsen, M. M'yllenborn, K. Birkelund and F. Grey, Appl. Phys. Lett., 69, 544 (1996); S. Sun, K.S.L. Chong and G.J. Leggett, J. Am. Chem. Soc. Comm., 124, 2414 (2002); J. Massanell, N. Garcia, and A. Zlatkin, Opt. Lett. 21, 12 (1996); G. Wysocki, J. Heitz, and D. Bäuerle, Appl. Phys. Lett. 84, 2025 (2004).
- [8] J. D. Beach, C. Veauvy, R. Caputo, R. T. Collins, A. A. Khandekar, T. F. Kuech, C. K. Inoki, T. S. Kuan and R. E. Hollingsworth, Appl. Phys. Lett. 84, 5323 (2004).
- [9] N. Kramer, H. Birk, J. Jorritsma, and C. Schonenberger, Appl. Phys. Lett. 66, 1325 (1995); S. C. Minne, Ph. Flueckiger, H. T. Soh, and C. F. Quate, J. Vac. Sci. Technol. B 13, 1380 (1995); N. Kramer, J. Jorritsma, H. Birk, and C. Schonenberger, J. Vac. Sci. Technol. B 13, 805 (1995).
- [10] M. K. Herndon, R. T. Collins, R. E. Hollingsworth, P. R. Larson, and M. B. Johnson, Appl. Phys. Lett. 74, 141 (1999).
- [11] H. A. Bethe, Phys. Rev. 66, 163 (1944).
- [12] M. N. Islam, X. K. Zhao, A. A. Said, S. S. Mickel, and C. F. Vail, Appl. Phys. Lett. 71, 2886 (1997).
- [13] A. H. La Rosa, B. I. Jakobson, and H. D. Hallen, Appl. Phys. Lett. 67, 2597 (1995).
- [14] T. W. Ebbesen et al., Nature 391, 667 (1998); H. F. Ghaemi et al., Phys. Rev. B 58, 6779 (1998); H. J. Lezec and T. Thio, Opt. Express 12, 3629 (2004).
- [15] P. D. Flammer, I. C. Schick, R. T. Collins, and R. E. Hollingsworth, submitted to Phys. Rev. Lett.

Author Biography

Reuben Collins received a B.A. in Physics and Mathematics from the University of Northern Iowa and M.S. and Ph.D. in Applied Physics from Cal Tech. He held positions as Research Staff Member, Manager of III-V Epitaxy, and Technical Consultant to the VP of Services Applications and Solutions at IBM Research. Since 1994 he has been a Professor of Physics and Director of the Center for Solar and Electronic Materials at the Colorado School of Mines.