Submicrometer Lithography by Near-Field Optical Microscopy

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ABSTRACT

Optimization of (i) intensity of illumination and (ii) thickness of resist was made looking for the conditions when high spatial resolution could be achieved by optical near-field lithography. Standard set-up of near-field illumination through a tapered Al-coated fiber tip was employed for the exposure of positive resist OFPR-5000(EG), which is photo-sensitive for wavelength $\lambda < 450$ nm. Tip was scanned along the line at near-field conditions of constant sample-to-tip separation to produce adjustable exposure dose of the spin-coated resist film. Femtosecond, 120 fs, pulses of the power $P < 1$ mW (at 82 MHz repetition rate) at 400 nm were coupled into a fiber (< 1 m length) and delivered to the surface of the resist for illumination.

The issues of NSOM fabrication using tapered Al-coated tips are addressed. To achieve a reproducible and high aspect ratio (approaching 1 : 1) NSOM-based lithography there should be found resists allowing to produce thin films (< 100 nm) with low surface roughness (< 10 nm).

Keywords: near-field optical microscopy, Al-coated silica tapered tips, lithography

1. INTRODUCTION

Current excimer laser lithography uses KrF laser, which operates at 248 nm and is achieving 130 nm resolution when imaging tricks such as phase shifting mask, oblique illumination are used in projection system with standard objective lenses possessing a numerical aperture of $NA = 0.7$. Further improvements along this direction are expected by the employment of ArF (193 nm) and F$_2$ (157 nm) lasers with an eventual aim to reach 80-90 nm resolution in the year 2005.¹ This challenge meets enormous difficulties in implementation due to the lack of appropriate optics (CaF$_2$, BaF$_2$ glasses are not of required quality) in VUV spectral region. Purified vitreous silica (ED-brand from Nippon silicas glass almost free of Cl and OH) is other candidate, since it has an extended transmission into UV region (up to 160 nm), but this silica is susceptible to the defect formation, too.²±³ Even the appropriate glasses would be finally obtained, their aging under the illumination of light, which quanta are comparable with the bandgap of those glasses eventually is expected to degrade the objectives (certainly, the employment of catadioptric objective lenses is other solution).

One of the alternatives to reach smaller feature sizes in lithography is near-field⁴ optical fabrication. This technique already has shown potential in the field where high light intensity is necessary to repair a photomask by ablation (micro-pipette tips were used).⁵ Here, we report on the investigation of the possibility to use Al-coated silica tapered tips to fabricate sub-$\mu$m features on the standard photo-resist by near-field scanning optical microscopy (NSOM).

2. EXPERIMENTAL

2.1. Self-made NSOM tips

NSOM fabrication was done using microscope Lumina, Topometrix (the fabrication area is shown in Fig. 1(a)) and commercial tips (Fig. 1(b)), which have been found differ between each other even in the same batch, or by self-made ones, what is specified in the text where it applies. In order to improve fabrication resolution we have paid attention on the geometry of tip’s apex and we have made our own tips to control the tips geometry and aperture...
size (Fig. 1(d,c)). This is crucial for reproducibility in lithography, since the tips need to be fresh made, otherwise the aperture is increasing due to oxidation $\text{Al} \rightarrow \text{Al}_2\text{O}_3$, by which Al is transformed into transmissive sapphire. The single-mode, $3.96 \mu\text{m}$ core ($125 \mu\text{m}$ diameter cladding layer) optical fiber 3M, Optical fiber FS-SN-3224 was pulled by Sutter Instr. Comp., P-2000 micropipette puller. The pulling force and CO$_2$ laser power were adjusted to obtain conical shape apex (Fig. 1(d)) and typically $1 \mu\text{m}$-length tip of reducing diameter. Then the tips were Al coated in an evaporator (Nilaco), which was modified to allow the rotation of fibers during the coating and, also, allowed their fixation at a pre-defined angle (Fig. 1(e)). The typical 90-100 nm Al coating was evaporated in one or two steps at the evaporation speed of 0.5 nm/s. Finally, the tips were inspected by scanning electron microscopy (SEM) on S-4200 (Fig. 1(c)). The aperture was recognized as darker (non-conductive) area on the apex of tip. There was no any observable degradation (on monthly scale) of the Al coating at the aperture edges, what also was confirmed by optical transmission measurements carried out using a Newport powermeter (10 pW precision). The fabricated tips were glued on the tuning forks (commercially available from Topometrix) using self-made 3D micro-stage and were further inspected for the resonance and aging in the circuit of the microscope Lumina. Finally the tips which had 100-150 nm diameter apertures were chosen for the NSOM lithography. The procedures of resist exposure were carried

**Figure 2.** (a) Absorption spectra (given as optical density (OD) vs. wavelength) of positive OFPR-5000(EG) resist at room illumination by a daylight (mercury) lamp of $9 \times 10^{-6}$ W/cm$^2$ (at $\lambda < 420$ nm) intensity after the different exposures: 1 - as made, 2 - 0.5 min, 3 - 3 min, 4 - 15 min, 5 - 25 min, and 6 - 40 min. The transmission profile of NSOM fabrication setup is given by curve filter. (b) Optical density (OD) vs. exposure time at 400 nm. Thickness of the film was 2.38 $\mu\text{m}$. The exposure $OD_{MAX} \times e^{-1}$ was reached after 120 min, what corresponds to the dose of 65 mJ/cm$^2$. 
Figure 3. (a) Surface topography of resist film after different exposures per line: from line 1 to 13 the exposures were 60, 1, 2, 3, 4, 5, 6, 10, 20, 30, 40, 50, 60 scans. One scan actually was a double (two-directional) exposure, which was totally collected over 5 s at a 4 μm/s scan speed. Line width was limited by a scanning step and was ca. 200 nm. The image is inverted, i.e. the illuminated lines were found with indentation. (b) Photobleaching (a decrease of transmission, \( -\Delta T \)) of resist under continuos NSOM exposure measured as photo-multiplier tube (PMT) voltage. The recharging of PMT after illumination is marked in the plot. (c) PL spectra recorded consecutively after every 10 s. The number of spectrum is given in the inset. The arrows show the tendency in spectral changes with exposure. Excitation was centered at 400 nm.

out on a modified NSOM microscope Lumina. The modifications were introduce to guarantee a stable feedback loop control, which keep a fixed tip-sample separation of ca. 10-15 nm. This was achieved by using lock-in amplifier in a feedback loop and windproof enclosure of all microscope to reduce vibrational noise. We employed sample (stage) scanning mode of operation to maintain constant optical axis and to avoid aberration-related distortions of illumination field.

2.2. Resist films and NSOM lithography procedures

The films of resist OFPR-5000(EG) were first diluted by acetone at 1:1 volume ratio, then, spin-coated over the cover glass using two-step procedure: 10 s at 600 rpm and then 20 s at 1000 rpm. After pre-baking at 110°C for 90 s the final thickness of films typically measured 1.35 ± 0.15 μm. Higher dilution and higher spin frequencies were employed to reduce the thickness of resist film, which was measured by profilometer Sloan DEKTAK-3030 and is specified where it applies. The roughness of the films was measured by atomic force microscopy (AFM) on

Figure 4. (a) Developed resist film after NSOM exposure. Line A was written in 68 scans, B - 34 scans, and C - 1 scan, where one line scan lasted 3 s at 1.5 μm/s. (b) The line written in 5 scans (3 s/scan ). Laser input power into the fiber was 0.1 mW (82 MHz laser repetition rate) at 400 nm. (c) An example of a developed line written by multi-scan irradiation (ca. 100 scans at 4 μm/s), which was approximately 5 μm-wide. Single scan illuminated line is visible in the developed film and measured ca. 1 μm-wide at the surface.
Figure 5. (a) An example of the line fabrication recorded by single scan in the 100-nm-thick resist film at 10 μm/s scanning speed when input laser power was 0.4 mW at 400 nm. Min-max height span was 91 nm. The cross-section A-A is given in (b). (c) Typical film’s roughness dependence on the film’s thickness. (d) The result of multi-line scanning (10 scans) in 300-nm-thick resist film at input power 0.1 mW (in the inset). Height cross-section was averaged over the rectangular depicted by dashed line.

Seiko Instr. SPI-300. Space-time-spectra resolved measurements were carried out on a modified near-field optical microscope Lumina, Topometrix.8

A NSOM light source was a second harmonic of Ti:sapphire laser (Tsunami, Spectra Physics) radiation at 370-420 nm wavelength with 120 fs pulses at 82 MHz (the cutoff frequency of fiber was 380 nm). Typical power of 0.1 mW was coupled into the fiber using a coupler with ×10 magnification objective lens (the laser power excess of 2 mW was degrading a tip’s aperture). The central 400 nm wavelength of illumination was at the edge of absorption band of resist (Fig. 2). The time lengthening of the pulse was about 30 times in ca. 1 m fiber as can be calculated by an empirical formula9:

\[ \tau_{out}(L) = \sqrt{1 + \left( 7 \times 68 \left( \frac{D L}{\tau_{in}} \right) \right)} \times \tau_{in} \]  

Here, \( D = 300; 450 \) fs/cm are the positive dispersions of fused silica and BK7 glass, respectively; \( L \) is the length traveled inside fiber, and \( \tau_{in, out} \) are the input and output duration of the pulse.

3. RESULTS AND DISCUSSION

The NSOM tip was scanned along the line at near-field conditions over the spin-coated resist film at a constant sample-to-tip separation of 10-20 nm in order to collect an adjustable exposure dose of resist film. As it was determined (Fig. 2) the photosensitivity (optical density (OD) dropped by exp(−1) times) of resist was ca. 65 mJ/cm² for 370-450 nm exposure at intensity 1.8 \( \times 10^{13} \) photons/(s·cm²). Corresponding exposure at near-field illumination needs to be collected over the repeated scans in a matter of seconds as can be evaluated from the typical fiber input power of 0.1 mW, pulse duration of 12 ps (a pulse spread due to group velocity dispersion (GVD) in fiber is taken into account by eqn. 1), tips throughput of 10⁻⁵, and tips aperture 100 nm was used for calculation. At theses conditions the sample is exposed to 5.8 kW/cm² intensity or 1.1 \( \times 10^{17} \) photons/s·cm² (or 2.5 photons per pulse at 82 MHz repetition rate). By knowing the photosensitivity of resist, the tip transmission can be calculated when illumination conditions are known. Such an evaluation gives tip’s transmission value of \( 10^{-4} \text{ } - \text{ } 10^{-5} \) in our experiments. An overall transmission from the fiber input down to the sample’s surface is very important and is usually not known ab initio due to the uncertainty in light losses at the fiber-tip interconnection.

The width of the lines exposed at NSOM conditions were found almost independent on the cumulative exposure (Fig. 3(a)). This should be expected if the photo-modification is following standard exposure law OD \( \propto \text{lg(Exposure)} \) rather than a photo-thermal diffusion law, since then the line-width of photo-modification is expected to follow Width \( \propto \sqrt{\text{Exposure}} \), which was not observed experimentally. The changes in optical density, \( OD \), of resist film occurred on the scale of seconds at near-field exposure (Fig. 3(b)). The consecutive photoluminescence, PL, spectra of the resist measured at near-field excitation, also, showed photo-degradation along with exposure (Fig. 3(c)).

After the exposure we have developed resist films to evaluate the dimensions of the exposed lines (Fig. 4). Typically few-μm-width lines were found in 0.6-1 μm-thick resist films at multi-scan exposure. This suggested to
consider the influences of scan speed, light wavelength and intensity, and resist’s thickness and roughness on the final width and depth of fabricated lines. It is obvious that a far-field component of the emanating light from the tip is exposing increasingly larger volume with the thickness of the film (a diffraction spread). The diffraction divergence of the Gaussian beam can be accounted for by the angle of diffraction $\theta$:

$$\theta = \frac{\lambda}{(\pi \omega_0)},$$

where $\omega_0$ is the waist (radius) of the beam at focus. We succeeded to fabricate the lines of sub-wavelength width when the thickness of resist was reduced to 100 nm (Fig. 5(a,b)). A decrease in the resist thickness can be done by strong dilution of the resist (the most efficient factor) and by larger velocities of spin coating. However, both approaches lead towards the increased roughening of the film and its decreased homogeneity (Fig. 5(c)). While the roughness on 20-25 nm scale was not a problem in multi-scan exposure when $\mu m$-wide lines were produced (Fig. 5(d)), it was found hampering the sub-$\mu m$ fabrication, since it is usually obtainable only by a single scan exposure (Fig. 5(a)). A projected thickness of the resist we used is 1 $\mu m$ (Sec. 2.2) and the thinning procedures still need to be optimized. From the data obtained so far, it is clear that the film thinning is the most efficient way to fabricate sub-wavelength features. Scanning speed of NSOM exposure was found not influencing the line width considerably (Fig. 6(a)), neither the wavelength. One more fact should be addressed, namely, a spectral widening of fs-pulses propagating in fiber due to self-phase modulation (SPM). Since SPM is nonlinear effect, i.e. light intensity dependent, the comparatively low intensities of the light pulses were causing minute spectral spreading, which can be neglected due to inherent 10-15 nm width of the pulses (at FWHM). However the SPM manifestation could be possible explanation of the line widening at input power of over 0.1 mW (the damage power of tip’s aperture was 2-3 mW). The Stokes shift caused by SPM can deteriorate the resolution at higher input powers (the anti-Stokes part is cut by the fiber bandwidth limitation at 380 nm).

The power dependence of the line width and depth was measured to investigate and to optimize the fabrication. As it can be seen (Fig. 6) the power dependencies of the fabricated line width and depth can differ from tip to tip. Two self-made tips with different aperture sizes of ca. 100 nm and 130 nm were utilized for NSOM exposure (Fig. 6(b)). If SPM and group velocity dispersion, which leads to the time spread of the pulses in fibers of different length according to eqn. 1, would define the exposure collected by resist, one could expect similar power dependence of the fabricated line width and depth, when the length of fiber, the input light power and the light-fiber in-coupling efficiency are the same. However, we did not observed any difference in fabrication for our used 0.5-1.5 $m$ fibers and the tips aperture was found to be decisive in the determination of the feature size (Fig. 6(b)). Even the tip scanning...
speed was not crucial for the width of the fabricated line (Fig. 6(a)). In fact, the tip's shape is influencing its Al coating, the final aperture size, heat dissipation during exposure, etc. The interplay of all these complex phenomena are finally deciding the fabrication quality and dimensions. As it can be seen (Fig. 6(b)), the resolution of the fabrication is deteriorating with the illumination power more significantly in the case of smaller aperture. This is direct consequence of diffraction, and the eqn. 2 is valid to explain the larger divergence from the smaller aperture.

4. CONCLUSIONS AND OUTLINE

Near-field optical fabrication is demonstrated and a NSOM-based photolithography can challenge the sub-μm range. The crucial parameters to achieve the constant reproducibility in fabrication are the control over the tip aperture and fiber apex shape together with the optimization of the resist itself to obtain as thin films as possible. For a higher resolution (to exploit real possibilities of NSOM) the following issues should be addressed:

1. Photoresist should be thin (< 150 nm) and flat (min-max roughness < 10 nm). This is uneasy to achieve by spin coating of OFPR-5000 resist due to high viscosity and bad solubility. Thin film would allow to avoid far-field component irradiation of the resist (this kind of far-field is highly divergent due to a diffraction spreading).

2. Higher sensitivity (to an optical exposure) of photoresist would allow to use a lower irradiation power, what would exclude a heating of the resist by Al coating of the tip's apex. Also, a faster fabrication time means shorter time of the tip in the proximity of the surface, where feedback control is sometimes failing to guaranty fixed tip-surface separation. This usually causes degradation of resolution as well.

3. NSOM fabrication would benefit from a higher transitivity of NSOM tip (milled Al-coated tips or pipette-type tips would be advantageous).

4. Continuos-wave or ns-pulsed lasers with narrow lasing line (1-10 cm\(^{-1}\)) are expected to increase the resolution. Especially promising is InGaN/GaN solid-state laser operating at ca. 400 nm used together with single mode fiber at the corresponding wavelength.

5. The procedures of chemical development need to be optimized in order to achieve minimum feature size.

REFERENCES