

Fabrication of large area 100 nm pitch grating by spatial frequency doubling and nanoimprint lithography for subwavelength optical applications

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In this article we report on the fabrication of 100 nm pitch gratings over a large area ($\sim 10 \text{ cm}^2$) using a simple, low-cost, fast process. This method includes (1) generation of the grating pattern using interferometric lithography and spatial frequency doubling and (2) pattern replication using nanoimprint lithography. The form birefringence of a 100 nm pitch Si grating was studied using ellipsometry. Measurements show an index difference of $\Delta n > 0.9$ at a wavelength of 632.8 nm. The experimental data are in good agreement with effective medium theory. This indicates the possibility of using these structures for wave plates and other subwavelength optical devices operating in the visible. © 2001 American Vacuum Society. [DOI: 10.1116/1.1409384]

I. INTRODUCTION

In the field of subwavelength optics, nanometer scale gratings have many important applications, such as in visible and infrared (IR) polarizing optical elements,^{1,2} extreme-ultraviolet (EUV) and UV transmission filters,^{3,4} broadband antireflection surfaces,⁵ phase retarders, and wave plates.^{6,7}

Many of the subwavelength devices are based on the effect of form birefringence⁸ which refers to the optical anisotropy arising from an ordered arrangement of optically isotropic materials on a scale much smaller than the wavelength of light.

In order to make compact and efficient subwavelength devices operating in the UV and visible, sub-100 nm pitch gratings are highly desirable. Previous fabrication methods include electron-beam lithography (EBL), atomic beam interference, and liquid immersion or achromatic interferometric lithography (AIL).^{9,10} However, they all have limited field size, require complex processing steps, and need expensive facilities.

In this article we report the fabrication of 100 nm pitch gratings over a large area using a simple, low-cost, fast process, which includes (1) fabrication of a 100 nm pitch grating mold by doubling the number of lines of a 200 nm period grating and (2) pattern replication using nanoimprint lithography (NIL).¹¹

Form birefringence of the 100 nm pitch grating was studied using ellipsometry. The experimental data are in good agreement with effective medium theory. The measurements show that, for a 100 nm pitch Si grating at a wavelength of 632.8 nm, a refractive index difference of $n_{\text{TE}} - n_{\text{TM}} > 0.9$ can be achieved. This indicates the usefulness of these gratings for a variety of subwavelength devices in the UV and visible.

II. FABRICATION

Currently, in the Nanostructure Laboratory at Princeton, we have developed technology that allows the fabrication of

large area (10 cm \times 10 cm) gratings with controlled line-widths and periods as small as 190 nm. The nanoscale gratings are generated using interferometric lithography (IL), with a continuous-wave (cw) Ar ion laser operating in single frequency mode as the light source. Subsequently, the pattern is replicated using a process combining NIL, liftoff, and reactive ion etching (RIE), with the interferometrically generated grating as the master mold.

However, the period of gratings generated by interference is limited by the wavelength of the light used for exposure. The minimum pitch obtainable by this technique using our exposure system is around 200 nm, which is about half of the Ar laser wavelength (351.1 nm) when the exposure is done in air.

To overcome this limitation, 200 nm period technology can be used in conjunction with a variety of edge defining techniques to double the number of lines of the original 200 nm period grating, effectively reducing the grating pitch by half.

A number of processing methods have been tested by us and have proved feasible. The schematic of one sequence of fabrication steps is shown in Fig. 1. It starts with the deposition of silicon nitride on a 200 nm period SiO₂ grating on a silicon substrate by low-pressure chemical vapor deposition (LPCVD). Second, the silicon nitride on top of and in the trenches between the grating lines was removed using CHF₃ RIE. Because the RIE etching is anisotropic, the sidewall silicon nitride remains and results in a grating pattern with half the original period after the SiO₂ is removed by a buffered HF wet etch. A 100 nm pitch grating mold can then be created in Si with the silicon nitride grating as an etching mask.

Once the master mold is obtained, grating patterns can be replicated by nanoimprint lithography using the steps shown in Fig. 2. In the NIL process, a layer of imprint resist was first spun on the substrate, and the grating mold was then pressed into the resist after the substrate was heated above the glass transition temperature of the polymer. After cooling, the mold was separated from the substrate, leaving a

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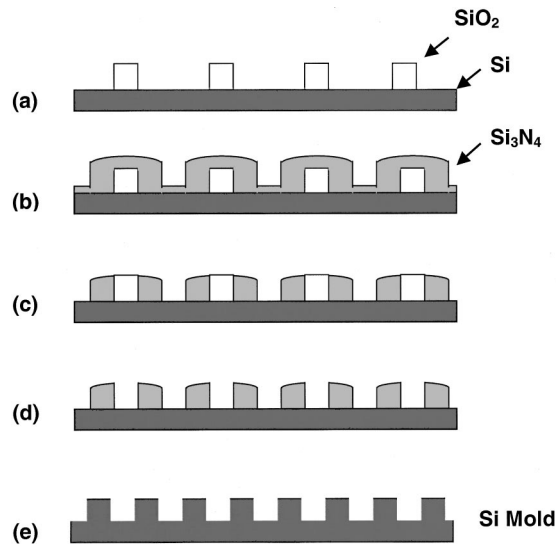


FIG. 1. Fabrication steps for a 100 nm pitch grating mold by doubling the number of lines in a 200 nm period grating: (a) The process starts with a 200 nm period grating in SiO_2 . (b) Deposition of CVD Si_3N_4 . (c) Reactive ion etching in CHF_3 and O_2 . (d) Removal of SiO_2 using diluted HF. (e) Pattern transfer into the Si substrate.

grating pattern in the resist. A number of subsequent processing steps can be used to transfer the pattern into the underlying substrate.

III. MEASUREMENTS AND ANALYSIS

For applications in subwavelength optical devices (polarizers, antireflection coatings, phase retarders, and wave plates), gratings with very fine lines and spacings are highly desirable. Many of the subwavelength devices are based on the effect of form birefringence, which requires linewidths and spacings that are sufficiently small compared to the wavelength, so the gratings can be treated as homogeneous, but also optically anisotropic, media.⁸

For low-index dielectrics, it has been shown that effective medium theory is usually a good approximation when the

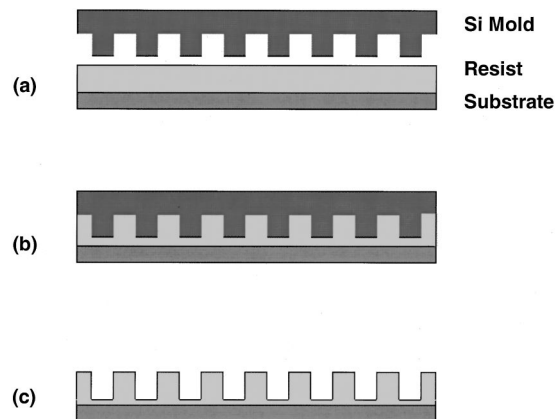
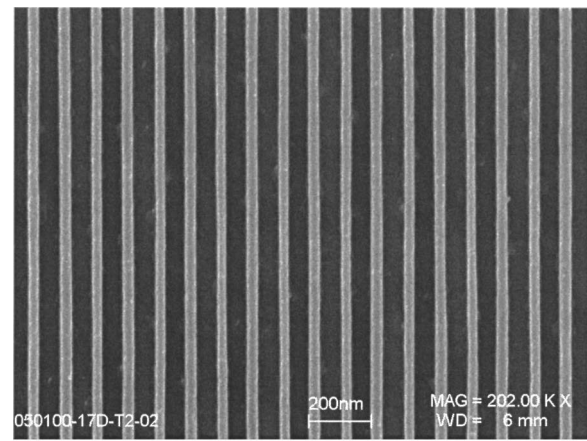
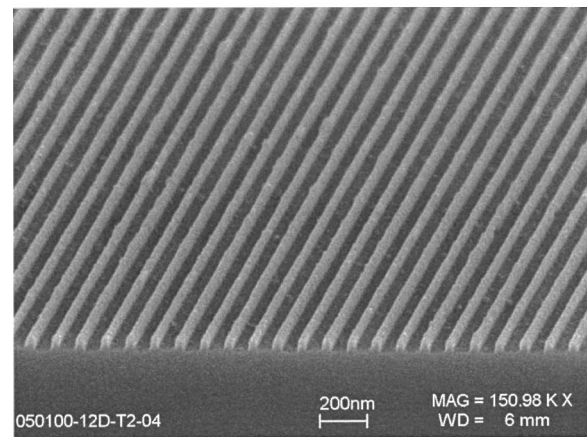


FIG. 2. Duplication of the 100 nm pitch grating using nanoimprint lithography: (a) The substrate is spin coated with a layer of imprint resist. (b) The grating mold is pressed in at a high temperature. (c) The mold is separated after the cooling process, leaving a grating pattern in the resist.



(a)



(b)

FIG. 3. Scanning electron microscope micrographs of a Si NIL mold with 35 nm linewidth and 2:1 aspect ratio fabricated by spatial frequency doubling showing deviation of less than 5 nm from perfect 100 nm periodicity: (a) top view; (b) cross section.

grating period is less than one half of the wavelength.¹² But for a high-index material such as Si, calculations indicate that, for the grating to be treated as an effective medium, a 100 nm grating period is generally required when operation in the visible range is desired.

Due to fabrication errors in spatial frequency doubling in the preparation of the mold, gratings fabricated using the method described in Sec. II are not perfect 100 nm period gratings. In general, the spatial frequency doubling process results in two different spacings between the grating lines. However, with tight process control, the difference between the two spacings can be less than 5 nm, as shown in Fig. 3.

It should be pointed out that form birefringence arises from the ordered arrangement of an optically isotropic material, but it does not directly depend on the period of the grating. As long as the linewidths and spacings are sufficiently small compared with the wavelength, and there is only one eigenmode propagating inside the grating, the effective indices (n_{TE} and n_{TM}) associated with this mode are independent of the grating period. Therefore, it is conceivable that small deviations from perfect 100 nm periodicity

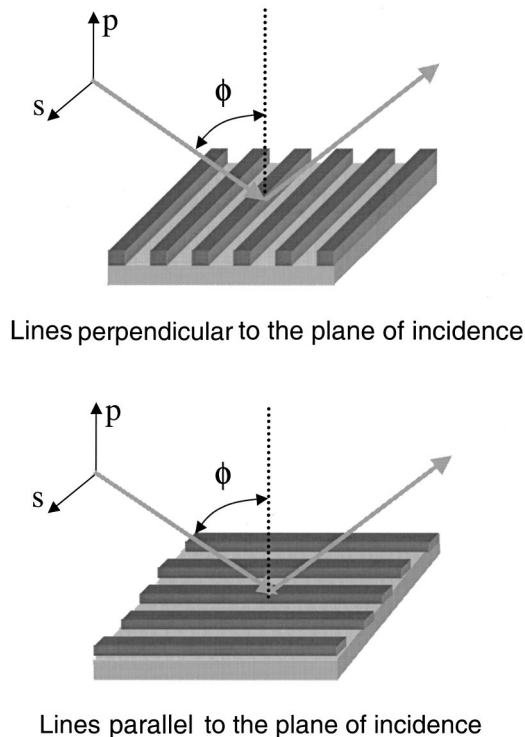


FIG. 4. Schematic of the orientation of the incident beam relative to the gratings in the ellipsometry measurement: The sample was measured at two orientations (shown by the lines perpendicular and parallel to the plane of incidence).

will not cause significant degradation in terms of form birefringence, and that the gratings fabricated using our method will be optically equivalent to a 100 nm period grating in that sense.

To test the usefulness of the 100 nm pitch gratings in subwavelength birefringent devices for the visible, optical properties of the Si grating shown in Fig. 3 (which has a linewidth of 35 nm and a 2:1 aspect ratio) were studied using ellipsometry at a wavelength of 632.8 nm. A schematic of the measurement is shown in Fig. 4. The ellipsometric parameters Ψ and Δ ($\tan \Psi e^{i\Delta} = R_p/R_s$, where R_p and R_s are the complex reflection coefficients of the p and s polarizations) of the beam reflected by the Si grating were measured at different angles of incidence, with the grating lines oriented perpendicular or parallel to the plane of incidence.

The measurement data and theoretical predictions based on the effective medium approximation^{8,13} are shown in Fig. 5. The difference between the data for different orientations of the grating relative to the plane of incidence is the result of a strong effect of form birefringence with $\Delta n > 0.9$ between the transverse electric (TE) (the electric field parallel to the grating lines) and transverse magnetic (TM) (the electric field perpendicular to the gratings lines) polarizations. The experiment data are also in good agreement with effective medium theory, which indicates these gratings can be used as artificial birefringent media for various subwavelength optical applications.

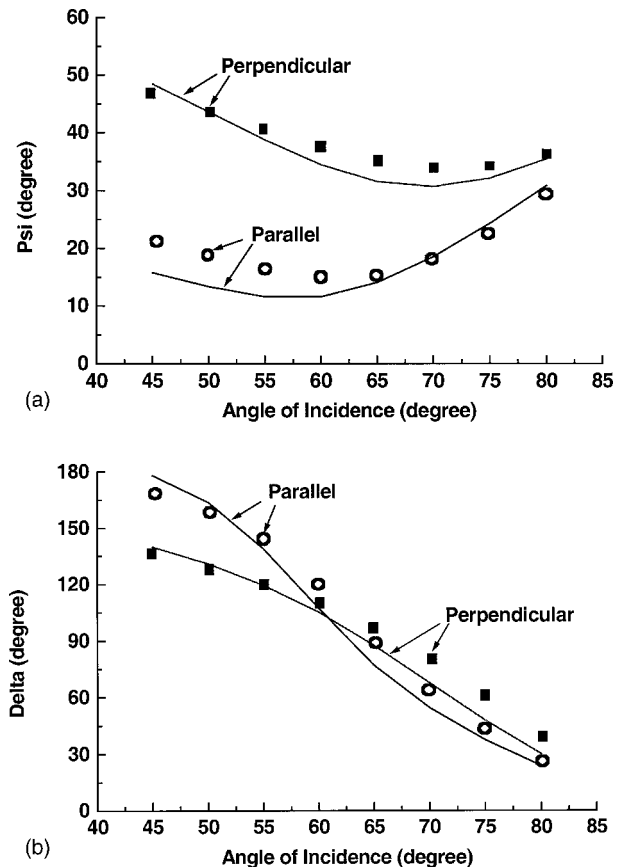


FIG. 5. Variation with the angle of incidence of the ellipsometric parameters Ψ and Δ ($\tan \Psi e^{i\Delta} = R_p/R_s$, where R_p and R_s are the complex reflection coefficients of the p and s polarizations) of the beam ($\lambda = 632.8$ nm) reflected by a 100 nm pitch Si grating. The experimental data (dots) are in good agreement with the simulation (lines) based on effective medium theory, which indicates a strong effect of form birefringence.

IV. CONCLUSION

Gratings with lines and spacings much smaller than the wavelength of light are highly desirable in the field of subwavelength optics. To make efficient subwavelength devices with high-index materials such as Si, 100 nm pitch gratings are generally required for operation in the visible. The combination of spatial frequency doubling and nanoimprint lithography can provide a simple, low-cost, fast method for the fabrication of such gratings over a large area. Because the lines and spacings are sufficiently small, their optical properties in the visible can be approximated by effective medium theory. These gratings can be used for a variety of subwavelength devices based on the strong effect of form birefringence.

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