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Laser interferometric nanolithography using a new positive chemical amplified resist

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The authors report on the progress of laser interference lithography at 266 nm laser wavelength with a chemical amplified resist containing a polyvinyl derivate dissolved in propylene glycol monoethyl ether ester. A continuous-wave deep-UV source combined with a Lloyd mirror is a simple and useful tool for the fabrication of nanoscale periodic structures generally called nanoarrays. Aiming for a robust pattern transfer technique to fabricate nanoarrays into magnetic materials, the authors investigated the utility of a chemical amplification positive tone resist, despite the relatively high theoretical resolution limit of 133 nm ($\lambda/2$) pattern period for the laser source used. Taking advantage of this new type of resist, the authors demonstrated for the first time the fabrication of an 18 Gbit/in.² dot pattern on a platinum thin film. © 2007 American Vacuum Society. [DOI: 10.1116/1.2800328]

I. INTRODUCTION

Current research in nanotechnology employs various nanolithographic methods. Techniques involved in these developments are state-of-the-art electron-beam lithography (EBL) employing 30–100 keV high-voltage sources, nanoimprint lithography, hot embossing, soft lithography, laser interference lithography (LIL), and hybrid lithography, which is the combination of different lithographic processes. EBL is a relatively slow direct-write method for periodic large-area nanostructures. It can additionally suffer from stitching and butting errors, for example, in optical grating.¹ Emerging lithographic techniques such as extreme UV, ion projection, and step-and-flash imprint lithographies are highly recognized for submicron patterning, but cannot be used as an initial patterning technique and will rely on a template or mask produced by another method. In most cases, EBL is used to produce these initial structures. EBL thus can be called a golden standard for pattern transfer at 20 nm lateral dimensions for the initial formation of arbitrary nanopatterns. Great advances have been made adopting an interferometrically controlled substrate table and advanced writing strategies using new, highly sensitive electron-beam resists. The limited throughput in the patterning process and high primary machine costs, nevertheless, remain a concern in numerous nanoapplications with reported throughput times per wafer up to 5 h.^{2–4} For the production of nanometer-scale periodic structures at a 100 nm pattern period, a very broad range of techniques is investigated and described in literature. For example, Solak and Ekinici have published on achromatic spatial frequency multiplication utilizing light produced at a synchrotron source.⁵ With respect to a full scientific comparison between the different nanofabrication techniques suitable for nanometer-scale periodic

structures, it is difficult to define an experimental design that covers important aspects such as throughput, minimum resolved features, and pattern density. These parameters are strongly dependent on machine performance, available access, and operator expertise. Comparing nanolithographic techniques is desirable for the development of an industrial roadmap, but is out of the scope of this work. Since our research focus is to study the magnetic structure of new nanoscale patterned media and the development of micro-electromechanical systems based recording systems, we use a LIL system with a commercially available laser source to fabricate patterned magnetic media test samples.^{6,7} A 6 nm bit cell spacing in patterned magnetic media with 1 Tbit/in.² has been the projected value elsewhere.⁸ Although this value is far beyond the capability of our LIL system, the technique has already been used by us for the investigation of patterned magnetic media, applying a cost efficient 266 nm deep-UV laser and standard i-line resists.⁹ This lithographic system provides a relatively cheap setup for the fabrication of nanoarrays surpassing the aforementioned restrictions of EBL with LIL exposure throughput times of about a minute or even less per in.², with dot pattern periods and diameters at the length scale of interest, for the investigation of patterned magnetic media data storage.⁹ To expand this recent work towards lateral dimensions at sub-50-nm, we introduce, here, a new chemical amplified positive tone resist (p-CAR) exposed by the Lloyd mirror interferometer setup. The optical arrangement is installed at the MESA+ NanoLab, which allows us to carry out nanopatterning in a class 1000 clean-room facility with a theoretical resolution limit producing lines and spaces at a pattern period of 133 nm ($\lambda/2$). In this paper, we define the lithographic process consisting of spin coating the new trilayer p-CAR resist system, deep-UV exposure and development. Preliminary studies on dry etch pat-

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FIG. 1. Nanolithographic process line at MESA+ Nanolaboratory. (A) Overview of the optical table $3 \times 3.60 \text{ m}^2$. (B) Shows the Lloyd mirror and the substrate holder in its relative size and position to the table. The inset has been made to receive a better visibility of the figure. (C) Shows the spin coater, which is dedicated to nanolithographic purposes, with the hotplate in operation. (D) Schematic of the optical setup.

tern transfer capabilities of the p-CAR resist are also discussed. Finally, LIL results are discussed in the context of patterned magnetic media.

II. EXPERIMENT

Figure 1(a) shows an overview of the LIL process line in the cleanroom. Figure 1(b) depicts the Lloyd mirror and substrate holder unit, which can rotate according to the angle of incidence (Θ), while the substrate can also be rotated around its own axis to carry out multiexposures at different substrate rotations (α). Figure 1(c) shows the spin coater in operation and a laminar flow wet bench for immersion bath development. A fourth harmonic continuous-wave yttrium aluminum garnet laser MBD 266 system (Coherent) is installed at the opposite side of the optical table (not shown in the figure). The laser beam is aligned and focused on a pinhole unit, which creates a Gaussian beam profile uniformly expanded towards the substrate holder over a distance of approximately 3 m for exposure of 4 in. wafers. Figure 1(d) schematically shows the optical setup. The new trilayer positive resist system used in this work consists of a stack of a bottom antireflection coating, BARC-DUV46 (Brewer Science), a photosensitive polyvinyl-based chemical amplified resist dissolved in propylene glycol monoethyl ether ester, p-CAR, commercially called PEK500 (Sumitomo Chemical), and a top antireflection coating, top coat Aquatar-6A (Brewer Science).

A. Dose profile modulation by BARC

For lithographic characterization experiments, semistandard polished 4 in. *p*-type silicon wafers were used as substrates. With the BARC thickness (t_{BARC}) as a variable parameter in the exposure experiments, the wafers were prepared by spin coating the resist system. Spin coating of all layers was performed on an OPTicoat ST22+ (Sister Semiconductor Equipment). Depending on spin speed between 1000 and 4000 rps/s, and coating condition working either with an open (OP) or closed (CL) coater chamber, we gained six different BARC thicknesses of 6 nm (CL4000), 13 nm (CL2000), 21 nm (CL1000), 29 nm (OP4000), 39 nm (OP2000), and 47 nm (OP1000), respectively. Spin conditions highly depend on the machine performance and must be optimized when a different machine for the coating process is used. Therefore, we do not suggest specific spin programs, but only the spin speeds in relation to each other. Solvent evaporation after spinning takes place on a closed hotplate in contact mode for all bake steps. The BARC layer was cured first for 30 s at 90°C , and then for 90 s at 205°C . To cool down the substrate, a chill step was performed by resting the substrate for at least 30 s on a plane metal surface at room temperature prior to applying the p-CAR. The photosensitive p-CAR was kept at a constant thickness of 140 nm during these initial exposure experiments. Softbake of p-CAR followed by a subsequent chill step was performed for 90 s at 110°C as described above prior to the application of the Aquatar. Aquatar is a very low viscosity material, and the spin parameters of Aquatar were kept constant (CL1000), resulting in negligible thickness. This layer does not require any bake steps. We did not investigate the impact of Aquatar spin conditions on the optical properties in the lithographic exposure, since the main function of this layer in our experiment is to seal the photosensitive layer from the ambient environment. The angle of incidence (Θ) defines the period (P) of the patterned array according to $P = \lambda / 2 \sin \Theta$, whereas λ is the exposure wavelength used, here 266 nm. All exposures were carried out at $54 \mu\text{W}/\text{cm}^2$ laser intensity. Subsequently, a postexposure bake of 90 s at 105°C was applied, and the resist was manually developed in an alkaline immersion development bath, OPD4262 (Fujifilm Electronic Materials), for 60 s under gentle agitation of the wafer. The wafers were rinsed in deionized water, first, by immersion and, secondly, by using a shower head. Subsequently, the wafers are spin dried, and cross-sectional samples were prepared. The samples were inspected by scanning electron microscopy (SEM) at fixed magnification using an xT-Nova Nanolab 200 dual beam system (FEI Company).

B. Pattern transfer by silicon dry etch

In this preliminary experiment, successful sub-50-nm lithography was carried out by performing a single exposure at $\Theta = 72^\circ$ ($P = 140 \text{ nm}$) using a reduced p-CAR thickness of 100 nm on a BARC thickness of 21 nm. The exposure time is 70 s. Bake and development steps remained the same as

for the previously described experiment. Dry etching was carried out in a parallel plate reactive ion etcher, PlasmaTherm 790 (Unaxis), running first a plasma etch step to break through BARC in an O_2 plasma [20 SCCM (SCCM denotes cubic centimeter per minute at STP)] at 100 mTorr, 100 W for 15 s. Subsequently, the silicon was etched in a standard fluorine plasma etch generated in $O_2(5 \text{ SCCM}):CHF_3(25 \text{ SCCM})$ at 30 mTorr, 350 W for 1 min. The etch receipt is generally optimized for etching silicon nitride on silicon, but was used in this work also for pattern transfer into BARC/silicon. This is possible when aiming for a relatively small etch depth, here 10–30 nm. After pattern transfer the remaining BARC can be stripped in oxygen plasma at 100 mTorr, 400 W for 1–2 min.

C. p-CAR LIL on sputtered platinum thin film

For this experiment, we prepared a 30 nm platinum (Pt) thin film by sputtering on a standard 4 in. silicon wafer. In thin films for data storage media, often a cobalt/platinum (Co/Pt) multilayer is used, whereas the top layer is a Pt layer. Since these Co/Pt multilayers are complex layers and generally structured by ion beam etching, we use in this work only Pt on silicon as a demonstration for the nanolithographic performance of the resist systems. Using lift-off, a stripe of Pt was removed across the wafer. This preparation method allowed us to compare the exposure and development performance on silicon against Pt in one process run. The wafer was processed in quarters. Dot arrays were produced by orthogonal double exposure (i.e., substrate rotation $\alpha=90^\circ$). The two subsequent exposures both were carried out for 21 s at $\Theta=45^\circ$ ($P=188 \text{ nm}$) using a resist thickness of 140 nm. The development was carried out in two steps, of which the first was 60 s and the second 30 s, respectively. The experiment was cross-checked with another quarter of the wafer, which was developed in two steps of 90 and 30 s, resulting in a total development time of 120 s. After each of the development steps, a sample was prepared for SEM inspection.

III. RESULTS

A new LIL nanolithographic process using p-CAR was installed at the MESA+ Nanolab according to Fig. 1 and evaluated. Figure 2 shows the results of a single exposure in the 140-nm-thick p-CAR layer at $\Theta=30^\circ$ ($P=266 \text{ nm}$) as a function of t_{BARC} using an exposure time of 28 s. Figures 2(b)–2(g) demonstrate how the linewidth to space ratio and side wall profile can be tuned in the p-CAR by the BARC layer thickness, leading to smallest footprints at 6 nm BARC. The BARC thickness thus can be used to vary bit cell spacing and dot diameter for the preparation of test samples in recording experiments. By taking the BARC layer of 21 nm, Fig. 3 presents successful sub-50-nm lithography in 100-nm-thick p-CAR at $\Theta=72^\circ$. Compared to the previous results depicted in Fig. 2 at $\Theta=30^\circ$, the exposure time needs to be increased to compensate for the loss of intensity due to the changed angle of incidence. We expect an increase of 2.8 according to $\cos(30^\circ)/\cos(72^\circ)$, which would result in an

Fig. 2. Optimization of p-CAR nanolithography at $P=266 \text{ nm}$. (A) SEM micrograph of p-CAR lines/spaces at CD 39 nm. (B)–(G) Shows the lithographic performance as a function of the BARC layer thickness, $t_{\text{BARC}}=6, 13, 21, 29, 39,$ and 47 nm from top to bottom, respectively, at the same SEM magnification as (A).

exposure time of approximately 78 s. Our experience, however, showed that this relation is not exact, and it was better to work with a slightly reduced exposure time, here 70 s. This dose correction is due to increasing reflectivity at higher angles of incidence. After exposure and development, also successful dry etching took place. Figure 3(a) depicts a cross section of the imaging layer on the BARC prior to the etch step. Figures 3(b) and 3(c) show the dry etch result of the lines/spaces pattern transfer at a critical dimension (CD) of 37 nm. Figure 3(b) coincidentally shows a cross section where the resist was partially covered by a particle during pattern transfer, allowing us to compare the remaining resist profile with the etched line profile in BARC/silicon. The resulting

Fig. 3. Pattern transfer etch test at $P=140 \text{ nm}$. (A) sub-50-nm p-CAR lines on 23 nm BARC/silicon, (B) Particle (left-hand side of image) protecting resist during etching, (C) Lines etched in BARC/silicon with dimensions.

FIG. 4. Cross sections of dot arrays in p-CAR on 21 nm BARC and $P=188$ nm. (A) dots on Pt/silicon after 60 s development, (B) on Pt/silicon after 90 s development, (C) on silicon after 90 s for comparison, and (D) optimized on Pt/silicon after 120 s development.

etch depth in silicon is approximately 10 nm. These experiments require further characterization, however, depicting the dry etch capabilities of the new resist system. In future work we plan to use this resist system also as a mask for chemically assisted ion beam etching into a magnetic material.

Highly uniform dot arrays are required for patterned media data storage. Although Fig. 2(b) depicts the smallest linewidth at CD down to 39 nm at 6 nm BARC thickness, however, we did not choose this value for further experiments on dot arrays. The footprint of a dot resulting from a double exposure at 6 nm BARC becomes too small to support the adhesion of the dot to the substrate. The exact dose profile deposited, and thus the resulting development profile of the individual dots into the imaging layer, is difficult to predict in maskless techniques such as interference lithography and requires further experimental as well as theoretical work. Therefore we chose a BARC of 21 nm, which produces structures with an approximate filling factor (linewidth/space ratio) of 50% as was depicted in Fig. 2(d). Our focus remained on the characterization of the resulting pattern in the imaging layer, which consequently can act as a mask for a diversity of etching processes. Figure 4 depicts a nanoarray of dots produced by an orthogonal double exposure for different steps in the development process. SEM inspection after the first step of development showed that the previously used development time of 60 s was not sufficient to open the structure [Fig. 4(a)]. Instead of adopting the exposure dose immediately, we continued the development for an additional step of 30 s. By increasing the development time thus to 90 s, the dots on silicon were overdeveloped [Fig. 4(c)], while they were still underdeveloped [Fig. 4(b)] on 30 nm sputtered Pt, which indicates the relatively strong influence of the substrate reflectivity on the accumulated intensity profile. Repeating the experiment on another quarter of the wafer directly developing for 90 s, the structures were again slightly underdeveloped, and we added another step of 30 s. This result, after a total of 120 s, shows a clean substrate surface with a dot pattern for further processing on the Pt layer [Fig. 4(d)]. A dot footprint diameter of 83.9 nm with a standard deviation of 2.2 nm has been achieved at a dot

FIG. 5. Overview of a 18 Gbit/in² dot array in p-CAR on 21 nm BARC and $P=188$ nm on Pt/silicon after 120 s development.

spacing of 198 nm, which yields a pattern density of 18 Gbit/in². This estimation of the diameter has been determined from the SEM calibration given in Fig. 4(d) across the inspected five dots. Figure 5 depicts an overview of the nanodot array on platinum.

IV. SUMMARY AND CONCLUSIONS

Using a p-CAR based on polyvinyl derivatives dissolved in propylene glycol monoethyl ether ester of a thickness of 140 nm on a BARC thickness of 21 nm, an 18 Gbit/in² dot array on a platinum thin film was achieved by laser interference lithography at a wavelength of 266 nm. We used a double exposure at $\Theta=45^\circ$, employing $54 \mu\text{W}/\text{cm}^2$ laser intensity for 21 s each with a postexposure bake at 105 °C for 90 s on a hotplate and 120 s development time in OPD4262. Further optimization of p-CAR LIL will allow us to process suitable test samples for patterned magnetic media, for example, by lift-off using the BARC as a sacrificial layer or direct patterning of a Co/Pt magnetic film using p-CAR as a mask in ion beam etching. The dot array achieved in the p-CAR imaging layer on Pt by LIL is the starting point for further developments in patterned media research and other fields where nanoarrays are explored. Optimizing the BARC thicknesses for each specific substrate surface can further improve the dot geometry of the resist, and thus, also influence the maximal dot uniformity and density using this nanolithographic technique for the fabrication of nanoarrays.

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