Bit-array patterns with density over 1 Tbit/in.² fabricated by extreme ultraviolet interference lithography

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(Received 10 June 2007; accepted 24 September 2007; published 7 December 2007)

The use of patterned magnetic media in future data storage devices requires the fabrication of two-dimensional periodic patterns with resolution well below 35 nm in terms of the pattern period. The authors have used extreme ultraviolet interference lithography based on transmission diffraction gratings to demonstrate arrays of structures in negative resists calixarene and hydrogen silsesquioxane. The produced patterns have density exceeding 1 Tbit/in.². Analysis of structure size and position on one of the patterns indicates variations of subnanometer magnitude. © 2007 American Vacuum Society. [DOI: 10.1116/1.2799974]

I. INTRODUCTION

The areal density of data storage on magnetic media found in hard disk drives has been increasing with a compound growth rate of between 40% and 100% per year since the early 1990s.^{1–3} Hard disk drives with areal density as high as 180 Gbits/in.² are currently in the market and units with density of more than 420 Gbits/in.² have been demonstrated in research laboratories of the industry (Seagate Technology). The increase in areal density has been achieved through relentless development efforts in many components of disk drives including read/write heads and thin film magnetic media. For the 180 Gbit/in.² generation, the area occupied by a bit is $\sim 60 \times 60$ nm² assuming a square shaped bit. The magnetic bits in such thin film crystalline media typically contain less than 100 grains. The number of grains in each stored bit has to be above a certain number to maintain an acceptable signal to noise ratio. Therefore as the bit sizes decrease, the grains must be made smaller as well. However, this scaling cannot be continued indefinitely because of the so-called superparamagnetic limit. This limit stems from the fact that as the volume of a grain shrinks so does the energy required to flip its magnetization. When this energy becomes comparable to the thermal energy ($k_B T$) at room temperature the magnetization of a grain can spontaneously change its direction. Such a spontaneous change of magnetization is of course not acceptable in a magnetic data storage device. This phenomenon is expected to limit the storage density on thin film magnetic media at some point between 500 and 1000 Gbits/in.².

A potential solution to this problem is to use patterned media where each bit is stored in a lithographically defined magnetic element (dot).^{1–3} The elements are fabricated to behave as single magnetic domains with a large enough volume so that they are able to maintain their magnetization at room temperature. Moreover they are isolated from each

other so that their magnetization can be individually switched and read. As an added advantage the transitions are expected to be sharper than continuous media. The magnetic elements should be placed in periodic arrays on circular tracks so that they can be accessed by a read/write head on a spinning disk.

A major obstacle to the realization of patterned magnetic media is the difficulty of mass producing the patterned substrates at an acceptable cost. The high areal density requirement above 500 Gbits/in.² necessitates dot sizes to be well below 35 nm (in terms of pitch). The size of the dots and spacing between them is also of critical importance for successful read/write operations. The adopted production technique has to be able to pattern large areas, with very high resolution periodic structures at an acceptable throughput and cost. The photolithography technology that has so successfully been used by the semiconductor industry is not expected to be a suitable solution to this problem because of its different requirements in terms of resolution and cost. Electron-beam lithography is capable of producing patterns with the required resolution but it faces significant challenges and limitations.⁴ First, the throughput limitations of this serial writing technique preclude its use in high volume fabrication of hard disks. Secondly, the current stop-and-go mode of operation found in most e-beam machines is not suitable for writing bit patterns on continuous circular tracks with the required accuracy because of inevitable field stitching errors. The solution to the first problem may be to write masters with e-beam and mass produce hard disks by replication with nanoimprint lithography (NIL). The second problem may be solved by the development of e-beam machines with rotary stages. However, maintaining the required bit placement accuracy on the order of 1 nm or less on a rotating stage is a nontrivial task. In addition, replication via NIL of such high-density bit patterns at high throughput needs to be addressed.

Extreme ultraviolet interference lithography (EUV-IL) has the potential for the fabrication of high-resolution bit-array

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FIG. 1. Cr diffraction gratings manufactured on Si₃N₄ membranes by e-beam lithography with periods of (a) 70 and (b) 40 nm.

patterns required for patterned magnetic media. It has recently been shown to be capable of creating 12.5 nm half-pitch line/space patterns.⁵ A variety of different structures including one-dimensional and two-dimensional patterns,⁶ and circular patterns⁷ have been demonstrated by EUV-IL schemes based on transmission diffraction gratings. Interference lithography experiments have been performed on both synchrotron and tabletop EUV sources.⁸ The short wavelength of EUV radiation and the practical absence of proximity effect make the achievement of nanometer scale resolution possible. Here we report fabrication of bit-array-type patterns with density over 1000 Gbits/in.² by EUV-IL.

II. EXPERIMENTAL DETAILS

Transmission diffraction gratings with periods between 40 and 70 nm were fabricated on Si₃N₄ membranes by e-beam lithography and reactive ion etching as described before (Fig. 1).⁹ The gratings were arranged in groups of four on the membranes to obtain four-beam interference patterns (Fig. 2). In configurations where all four gratings have the same period the relative phases of the gratings have a profound impact on the interference pattern.⁶ We controlled the phases in the e-beam writing step to obtain two different kinds of

FIG. 2. Layout of a diffraction grating mask for four-beam interference lithography. When such a mask is illuminated at normal incidence by a spatially coherent beam, diffracted beams originating from the four gratings overlap at a certain distance from the mask and form an interference pattern. The horizontal and vertical gratings may have different periods, p_1 and p_2 as shown in the layout.

FIG. 3. SEM images of dot-array patterns in calixarene resist after development in xylene. The periods of the patterns are (a) 49.5, (b) 42.4, and (c) 35.4 nm. The scale bars are 100 nm long.

interference patterns that facilitate the fabrication of either dots or holes in a negative tone resist. We also experimented with cases where two of the gratings (parallel to each other) had one period and the other two (perpendicular to the first two) had a different period. This leads to an incoherent summation of two interference patterns due to the limited temporal coherence of the EUV light that we used.¹⁰

The EUV experiments were performed at the XIL beamline of the Swiss Light Source. This beamline delivers a spatially coherent EUV beam from an undulator source. The central wavelength of the illuminating beam was 13.4 nm and it had a spectral width of about 2%–3%. Two different negative tone resists, a calixarene resist (*p*-chloromethyl-methoxy-calix[4]arene, TEBN-1 from Tokuyama Corp., Japan) and a hydrogen silsesquioxane (HSQ) resist (FOX12 from Dow Corning) were used in the EUV exposures. Calixarene was spin coated onto Si wafers as received from the supplier, resulting in a film thickness of 25 nm. The samples were baked on a hot plate at 110 °C for 60 s after coating. The exposed samples were developed either in isopropyl alcohol (IPA) for 30 s or in xylene for 5 s. We had previously measured the EUV sensitivity of this resist to be 120 mJ/cm² when it is developed in IPA.⁵ Development in xylene reduces the sensitivity by a factor of about 1.5.¹¹

HSQ was diluted by adding methyl isobutyl ketone to obtain a film thickness of 35 nm. The samples were baked at 90 °C for 180 s after coating. Exposed samples were developed in 2.6*N* tetramethylammonium hydroxide (TMAH) solution for 10 min. The measured EUV sensitivity of HSQ under these processing conditions is 24 mJ/cm².¹²

FIG. 4. SEM images of dot-array patterns in calixarene resist after development in IPA. The periods of the patterns are (a) 31.8 and (b) 28.3 nm. The scale bars are 100 nm long.

III. RESULTS AND DISCUSSION

When the phases of the four interfering beams are controlled so that all four add up in phase we obtain a square array of intensity peaks which are surrounded by broad regions of intensity nulls.⁶ The period of this lattice is equal to the period of the diffraction gratings divided by square root of 2. Dot arrays with periods in the 49.5–28.3 nm range shown in Figs. 3 and 4 were recorded in calixarene using such interference images. The data storage density corresponding to these arrays is between 263 and 806 Gbits/in.². The smallest dots seen in Fig. 4(b) are about 12 nm in diameter. The samples in Figs. 3 and 4 were developed in xylene and IPA, respectively. Development in xylene offers the advantage of higher contrast but swelling in this developer sometimes leads to a collapse and merging of the produced structures.^{11,13} Dot arrays obtained in HSQ by this exposure method with periods of 49.5 and 42.4 nm are shown in Fig. 5. Higher-resolution exposures in HSQ did not produce isolated dot arrays due to remaining resist material between the dots.

Hole arrays were obtained by either controlling the phases of four gratings with identical periods or by using different periods in the two orthogonal directions. The pattern shown in Fig. 6(a) was obtained by using a mask that had four gratings all of which had 50 nm period. The phases of the gratings were written by e-beam so as to obtain an interference pattern that is equivalent to incoherent addition of two two-beam interference patterns perpendicular to each other.

FIG. 5. SEM images of dot-array patterns in HSQ. The periods of the patterns are (a) 49.5 and (b) 42.4 nm. The scale bars are 100 nm long.

FIG. 6. Hole-array patterns in calixarene. (a) A square array with 25 nm period. (b) Array with periods of 30 and 35 nm in the vertical and horizontal directions, respectively.

This interference pattern is suitable for obtaining hole arrays in a negative resist because of the presence of intensity nulls that are isolated from each other. This interference pattern has a lattice constant that is equal to half of the grating period in both directions. Therefore the hole array seen in Fig. 6(a) has a period of 25 nm with a corresponding data storage density of 1032 Gbits/in.². We also experimented with asymmetric arrangements where gratings in the horizontal and vertical directions had different periods. This method too leads to an image that is equivalent to an incoherent addition of two two-beam interference patterns. The image in Fig. 6(b) was obtained with a mask that had 60 and 70 nm period orthogonal gratings. The resulting hole array in resist has 30 and 35 nm periods in the vertical and horizontal directions, respectively, with a corresponding density of 614 Gbits/in.². Both samples seen in Fig. 6 were developed with the help of ultrasonic agitation in IPA for 30 s to help with clearing of the holes.

As we mentioned before the uniformity of the size and the spacing between the dots are critical for patterned magnetic media.³ We analyzed some of the images using commercial software to quantify the variations in dot size and spacing. The results are summarized in Table I for the analysis performed on the scanning electron microscopy (SEM) image in Fig. 3(b). The dot spacing analysis was done along four different directions indicated in Fig. 3(b). The standard deviation in both size and position is found to be around 0.5 nm for this pattern. First, we point out that this variation in size is in essence similar to surface roughness found in many spin-coated and annealed organic films such as photoresists. In fact, this variation is on the same order as the size of one calixarene molecule,¹⁴ indicating the achievement of control at the molecular size level. This result marks the achieve-

TABLE I. Analysis results for the dot-array image shown in Fig. 3(b) along the directions shown in the same image.

	Average (nm)	σ (nm)	σ /average (%)
Direction A, dot-to-dot distance	59.9	0.5	0.9
Direction B, dot-to-dot distance	59.9	0.5	0.8
Direction A', dot-to-dot distance	42.7	0.6	1.4
Direction B', dot-to-dot distance	42.0	0.5	1.3
Dot size	25.8	0.4	1.7

ment of subnanometer control in fabrication of two-dimensional structures obtained via top-down engineering means. The nature of interference lithography favors the uniformity of size and position. In our diffraction grating based interference scheme each point in the image receives contribution from a certain area on each of the four gratings. This is in contrast to a projection lithography system where there is a one-to-one correspondence between points in the object (mask) and the image. A comparison of the images of diffraction gratings in Fig. 1 and images of dots in Figs. 3–5 demonstrates that the EUV-IL scheme averages over large enough areas of the gratings to eliminate the effects of significant linewidth and position variations present in the gratings. Interference method has also an advantage over e-beam lithography as it is not susceptible to momentary disturbances in position and electromagnetic fields. It is also instructive to consider the effects of shot noise in the EUV exposure of dots. For the dots in Fig. 3(b) we have estimated the number of absorbed photons for each dot to be around 7000. Therefore we can estimate the relative statistical deviation in the number of photons exposing each dot to be 1.2%. While this number is similar to the experimentally found dot size variation, a direct comparison is difficult to make because one needs to consider the three-dimensional distribution of absorption events and their impact on size variation. For example, a photon absorbed near the center of a dot may not contribute to the size variation as much as one absorbed near the edge. Molecular level simulation of absorption events and their impact on dissolution and eventual formation of the resist structures upon drying may provide an answer to this question.

IV. CONCLUSIONS

We have recorded two-dimensional bit-array-type patterns at areal density levels relevant for the fabrication of patterned magnetic media for data storage in two different types of negative resists. The ultimate limit for resolution available by EUV-IL is equal to about 7 nm in terms of the pattern period that corresponds to a storage density in excess of 13 Tbits/in.². While many issues such as the need for resists

with the required resolution need to be addressed, the practical absence of proximity effect at the EUV energy may allow us to approach this limit in the future. The variation in size and spacing of the dot arrays is well below 1 nm. This unprecedented result obtained with a top-down patterning method rivals other methods such as chemical synthesis and assembly in terms of the monodispersity of the produced dots. The diffraction gratings that are used in the formation of the interference patterns can be manufactured with circular shape to make the bit-array patterns along circular tracks.⁷

ACKNOWLEDGMENTS

The authors thank Justin Hu, Kim Y. Lee, David Kuo, and Dieter Weller of Seagate Technology for performing the size and spacing variation analysis on resist images. They also thank Laura Heyderman for many useful discussions on patterned magnetic media. Part of this work was performed at the Swiss Light Source, Paul Scherrer Institute, Villigen, Switzerland.

¹B. D. Terris and T. Thomson, *J. Phys. D* **38**, R199 (2005).

²R. F. Service, *Science* **314**, 868 (2006).

³H. J. Richter *et al.*, *Appl. Phys. Lett.* **88**, 222512 (2006).

⁴S. Hosaka, H. Sano, M. Shirai, and H. Sone, *Appl. Phys. Lett.* **89**, 223131 (2006).

⁵H. H. Solak, Y. Ekinici, P. Kaser, and S. Park, *J. Vac. Sci. Technol. B* **25**, 91 (2007).

⁶H. H. Solak, C. David, J. Gobrecht, L. Wang, and F. Cerrina, *J. Vac. Sci. Technol. B* **20**, 2844 (2002).

⁷H. H. Solak and C. David, *J. Vac. Sci. Technol. B* **21**, 2883 (2003).

⁸P. W. Wachulak, M. G. Capeluto, M. C. Marconi, C. S. Menoni, and J. J. Rocca, *Opt. Express* **15**, 3465 (2007).

⁹H. H. Solak, C. David, J. Gobrecht, V. Golovkina, F. Cerrina, S. O. Kim, and P. F. Nealey, *Microelectron. Eng.* **67–68**, 56 (2003).

¹⁰H. H. Solak, *Microelectron. Eng.* **78–79**, 410 (2005).

¹¹M. Narihiro, K. Arai, M. Ishida, Y. Ochiai, and Y. Natsuka, *Jpn. J. Appl. Phys., Part 1* **44**, 5581 (2005).

¹²Y. Ekinici, H. H. Solak, C. Padeste, J. Gobrecht, M. P. Stoykovich, and P. F. Nealey, *Microelectron. Eng.* **84**, 700 (2007).

¹³S. Yasin, D. G. Hasko, and F. Carecenac, *J. Vac. Sci. Technol. B* **19**, 311 (2001).

¹⁴M. Ishida, J. Fujita, T. Ogura, Y. Ochiai, E. Ohshima, and J. Momoda, *Jpn. J. Appl. Phys., Part 1* **42**, 3913 (2003).