I. INTRODUCTION

Zone plates are the optical key elements in high resolution x-ray microscopy. The zone plate objectives have typically diameters of about 100 μm and several hundred zones. For short exposure times in x-ray microscopes and/or minimum radiation load on the specimen the diffraction efficiency has to be maximized for the zone plate objective. The achievable spatial resolution \( \delta \) of an x-ray microscope is determined by the outermost zone width \( d_{rn} \) of the zone plate objective. According to the Rayleigh criterion, we obtain \( \delta = 1.22d_{rn}/m \), where \( m \) denotes the order of diffraction which is used for imaging. For high spatial resolution, either the outermost zone width has to be fabricated as small as possible, zone widths down to 15 nm have been reported, or a higher order of diffraction \( m \) has to be applied.

The application of higher orders leads usually to too low diffraction efficiencies, since the efficiency scales with \( 1/m^2 \) for conventional zone plates. For the so-called volume zone plates with very high aspect ratio structures and tilted zones this rule is no longer valid. This type of zone plate is highly efficient in a chosen high order of diffraction. The advantage of this kind of diffractive optics is that for high spatial resolution and high efficiency only a moderate zone width has to be fabricated but at the cost of generating very high aspect ratios and tilted structures. However, the required aspect ratios combined with tilted zones cannot be generated by existing nanostructuring processes for zone plate fabrication.

By applying the first order of diffraction for imaging, the efficiency of high resolution hard x-ray zone plates is limited by the achievable aspect ratios of the nanostructures. The diffraction efficiency strongly depends on the height of the diffracting zone structures. In general, the height of the zone structures for high efficiency values is significantly larger than the outermost zone width \( d_{rn} \). Therefore, high aspect ratio zone structures have to be fabricated. Furthermore, the required zone height strongly increases with the x-ray photon energy. It should be pointed out, that the highest achieved aspect ratio so far for zone plates with 20 nm outermost zone width is about 9:1. For example, the required aspect ratio for nickel zones with 20 nm outermost zone width with a first order diffraction efficiency of 22% is about 10:1, 25:1, and 50:1 for 0.5, 2, and 5 keV photon energy, respectively. For higher photon energies gold is often the preferred zone plate material, since the required aspect ratio is slightly lower compared to nickel. In the case of gold zone plates, for 2 and 5 keV photon energy and 22% diffraction efficiency aspect ratios of about 20:1 and 35:1 are required, respectively. This is out of the range of currently used nanotechnology processes. Therefore, in practice the performance of zone plates is a trade-off between obtainable resolving power and diffraction efficiency.

In this article, we propose to superimpose zone plates on top of each other on the same wafer to overcome the aspect ratio limits. With this approach, the aspect ratio is only limited by the number of stack layers. Figure 1 shows a calculation of the first order diffraction efficiency as a function of the photon energy and the number of layers. The zone plate material is nickel and the thickness of each layer is 200 nm. The calculation was performed for a zone plate with 20 nm outermost zone width and nontilted zones using electroly-
Summary of the document:

Electrodynamic theory.6,8 The aspect ratio of the structures in a single layer of the stack is 10:1 and the total zone height is given by the number of layers times the layer height of 200 nm. For the different photon energies the number of layers is increased, i.e., is plotted until the maximum efficiency is reached. At 0.52 keV photon energy the maximum is already reached for a single layer and no superimposing of zone plates is required. At higher photon energies, the efficiency can be significantly increased for each layer which is added to the proposed stack of zone plates. For example, at 2 keV photon energy superimposing of two zone plates (two layers) is increasing the efficiency from 6.6% (one layer) to 18% (two layers). Figure 2 shows for comparison a corresponding plot for gold as zone plate material. The maximum efficiency at 10 keV photon energy is obtained for a lower number of layers compared to nickel. At 5 keV the maximum efficiency of 23.5% is obtained with five gold layers which is comparable to the efficiency of 22% in case of five nickel layers. However, higher efficiencies are possible using nickel instead of gold (35% with nine layers). With three layers at 2 keV photon energy the maximum efficiency of about 24% is equal for gold and nickel zones.

For the successful development of stacked zone plates, two key technologies are of great importance: electron beam lithography with high overlay accuracy and the planarization of each single zone plate layer after electroplating. Recently, it was reported that the required overlay accuracy in the nanometer range can be achieved by electron beam lithography.3 However, the second required step of polishing zone plates was unsolved so far. Although chemical-mechanical polishing (CMP) processes for nickel and nickel based alloys have been explored9–11 and used for planarizing nickel nanostructures,12,13 they have not been applied in zone plate processing. We report here on the first results of polished nickel zone plates which is an important step toward the realization of stack zone plates with extremely high aspect ratios and as a consequence very high diffraction efficiency.

II. PROPOSED PROCESS FOR STACK ZONE PLATES

The proposed stack zone plate process is illustrated in Fig. 3. The suitability of the layer system is already demonstrated in conventional zone plate fabrication techniques.7,14,15 In...
practice, the processing steps 2–4 are under control. However, it is very difficult to fill completely the mold by electroplating and to avoid overplating which results in a craggy surface no longer following the shape of the zone structures. One approach to avoid this problem is to measure in situ the transmitted visible light through the zone plate during plating and to stop the deposition exactly at the top edge of the mold. This method cannot be used for the proposed stack process where several zone plates have to be fabricated on top of each other because the transmission of visible light through the stack is already too low during plating of a second zone plate on top of a first zone plate.

Our new approach to solve the electroplating problem is CMP of the rough surface after purposely overplating the mold. As shown in the processing step 4 of Fig. 3 the mold is overplated to exploit the maximum aspect ratio given by the plating mold. Afterwards the overplated nickel is removed by a CMP process (see Fig. 3, processing step 5) and a new layer system is added on top of the polished zone plate pattern (see Fig. 3, processing step 6).

In a second electron (e)-beam lithography step a zone plate pattern is exposed into the resist well aligned with the nickel zone plate underneath. This overlay exposure has to be done by using alignment marks which are located on the substrate (not shown in the illustration). The overlay of the zone plate patterns has to be very accurate, the overlay error should be less than $2dr_p/7$, whereas $dr_p$ denotes the outermost zone width. Recently an overlay accuracy of about 2 nm was reported, which demonstrates that the required overlay accuracy is possible. After the overlay exposure the pattern is developed and transferred into the second polymer layer by reactive ion etching (RIE) (see Fig. 3, processing step 7) as described for patterning the first zone plate. A second plating (see Fig. 3, processing step 8) and polishing step (see Fig. 3, processing step 9) is performed to obtain the final second nickel zone plate layer. By fabricating the second zone plate layer on top of the first zone plate layer the aspect ratio of the structures can be doubled. If the aspect ratio has to be increased further, additional layers can be added on the zone plate stack (see Fig. 3, processing step 10). Finally, the polymer mold will be removed by an O$_2$ RIE step (see Fig. 3, processing step 11). For the soft x-ray range the zone plate has to be on a thin support membrane, usually a 50–100 nm thick silicon nitride (Si$_3$N$_4$) membrane. Since the CMP process can only be applied on a thick substrate, the substrate has to be thinned after the final zone plate fabrication.

### III. CHEMICAL-MECHANICAL POLISHING OF NICKEL ZONE PLATES

Polishing each zone plate layer is vital for the success of the proposed stack zone plate process. CMP is a process of smoothing and planning surfaces of two different materials with the combination of chemical and mechanical forces, a hybrid of chemical etching and free abrasive polishing. Mechanical grinding alone causes too much surface damage, while wet etching alone cannot attain good planarization. CMP was first introduced in semiconductor fabrication in the mid 1980s. Since that time, CMP has emerged as the preferred manufacturing process for planarizing submicron multilevel metal layers in integrated circuits. We performed experiments to planarize nickel nanostructures embedded in a polymer mold by using a commercial polishing pad and a polishing slurry model MSW 1500 which is primarily designed for tungsten CMP. It has been reported that this slurry provides good results for planarizing FeNi microstructures in a SU-8 mold which is a similar material combination used for producing electroplated nickel zone plates. We used ARC-XL20 from BREWER SCIENCE as the polymer mold material. The hardness of the mold was significantly improved by crosslinking with x rays using an x-ray dose of about $2.5 \times 10^9$ Gy. After crosslinking the polymer has sufficient mechanical stability for CMP. Since the crosslinked polymer is chemically inert polishing of the polymer is a pure mechanical process. It has been observed that the adhesion of the polymer is sufficiently high to avoid separation of the polymer mold from the substrate. For the zone plate fabrication we coated a silicon wafer substrate (500 μm thick) with a 5 nm thick chromium and 10 nm germanium layer, which forms the conductive plating base for an electroplating process. Afterwards an ARC-XL20 layer was spin coated and crosslinked by x rays. After crosslinking the ARC-XL20 thickness was 175–200 nm, depending on the sample batch. Finally a 3.5 nm thick titanium (Ti) layer and a 40 nm thick e-beam resist layer (ZEP7000) was deposited onto the ARC-XL20 to obtain the final layer system as illustrated in Fig. 3, step 1. Zone plate patterns with 40, 30, and 25 nm outermost zone width and diameters of 62, 54, and 66 μm were exposed into the resist by e-beam lithography (Leo1560 scanning electron microscope with NANOMAKER lithography attachment). After development the pattern was transferred into the Ti layer using RIE with a BCl$_3$ plasma (see Fig. 3, processing step 2). The structured Ti layer serves as an etch mask for structuring the polymer layer during an oxygen plasma RIE step to obtain the final plating mold (see Fig. 3, processing step 3). The mold is filled by electroplating using a nickel sulfamate bath (Lectro-nic 10–03s, Enthone Inc.). The zone plate mold was about 50–70 nm overplated (see Fig. 3, processing step 4). The CMP process was used to polish the zone plate until the overplated nickel was removed (see Fig. 3, processing step 5). To judge if the overplated nickel is removed the CMP process was interrupted, the zone plate was cleaned in water and isopropanol to detach the residual slurry and investigated under a light microscope in differential interference contrast (DIC) mode. In case that overplated nickel was already removed a characteristic cross pattern in the zone plate becomes visible (see the DIC image of the polished zone plate in Fig. 7). The pattern results from the interaction of the circular, quasiperiodic structures of the zone plate with the polarized light from the microscope. If the cross was not visible, indicating that the overplated nickel was not removed, the CMP process was continued until the cross became visible. Finally, the zone plate was investigated in the scanning electron microscope to
cross-check if the overplated nickel is completely removed. After the successful polishing of the zone plate the plating mold was removed. For the removal an oxygen plasma RIE step is sufficient, since the Ti mask was already removed during the polishing step.

**IV. RESULTS**

A scanning electron microscope (SEM) micrograph of a polished nickel zone plate with 40 nm outermost zone width is shown in Fig. 4. The plating mold was removed by RIE in an oxygen plasma. In the upper micrograph of Fig. 4 the innermost part of the zone plate, in the lower image the outermost 40 nm wide zones of the zone plate are shown. The height profile of the polished nickel zone plate was measured with a line scan profilometer (Tencor P15). We measured for the outermost zones a height of 156 nm and for the innermost zones a height of 172 nm. The initial plating mold height was 175 nm. Note that small scratches are visible in the nickel structures. However, their depth is only a few nanometers and they will not degrade the zone plate properties.

We also demonstrated that the CMP process can be applied to smaller zone widths as shown in the SEM micrographs (see Fig. 5, lower SEM micrograph and Fig. 6) of polished nickel zone plates with 30 and 25 nm outermost zone width, respectively. No shifting of the zones or adhesion problems due to the CMP process were observed. The initial plating mold was 200 nm thick and overplated by about 60 nm. The overplated 30 nm wide nickel zones are shown in the upper SEM micrograph of Fig. 5. The lower micrograph in Fig. 5 shows the 30 nm wide zones after the CMP process and the removal of the plating mold by RIE.

We exposed an additional 7 μm wide ring surrounding each zone plate. The inner ring radius is about 3 μm larger than the zone plate radius. This ring pattern was used for accurate measurement of the plating mold height close to the zone plate pattern by using the stylus profilometer after the oxygen etch step (see Fig. 3, processing step 3). For the profile measurement the wide ring pattern was used, since

---

**Fig. 4.** SEM micrographs of a polished nickel zone plate with 40 nm outermost zone width (45 degree tilted view). Top: Innermost part of the zone plate. Bottom: Outermost part of the zone plate.

**Fig. 5.** SEM micrograph of the outermost part of a nickel zone plate with 30 nm zone width. The upper image shows the overplated 30 nm wide outermost zones before polishing and the lower image the 30 nm wide zones after polishing.

**Fig. 6.** SEM micrograph of polished 25 nm wide outermost zones.
the stylus tip radius is too large compared to the zone widths to measure the polymer zone height directly within the zone plate pattern.

Figure 7 shows profilometer line scans of a nickel zone plate with 30 nm outermost zone width after overplating (black line) and after polishing and removing the plating mold (gray line). The scan path is illustrated in the inserted light micrograph (DIC contrast) which shows the polished nickel zone plate and the surrounding nickel ring. Note that the line scans across the zone plate show two broad peaks which correspond to the ring pattern. The upper scan (black line) shows the height profile of the overplated nickel zone plate which is still embedded in the plating mold. An offset of 200 nm was added to this scan in order to take into account the plating mold height. One can clearly reveal that the zone plate is overplated (50 nm in the outer part up to 75 nm in the inner part), since the grown nickel structures are higher than the surrounding mold material. The inhomogeneous nickel height is due to an observed effect that the plating rate depends on the linewidth, i.e., the aspect ratio of the structures. The rate is lower in the outermost structures with their high aspect ratios. The line scan below (gray line) shows the height profile of the nickel zone plate after polishing and removing the plating mold. The final structure height ranges from about 150 nm in the outer zone plate area to about 180 nm in the inner zone plate area. A similar height distribution was already present before polishing, which means that the height distribution of the nickel zone plate was not much affected by polishing. The loss in mold material thickness which was initially 200 nm is about 20–50 nm. We expect that a more homogeneous height distribution of the nickel plating automatically improves the height distribution of the polished nickel zone plate. We assume that pulsed plating solves this problem.

The authors gratefully acknowledge H. H. Gatzen and G. Günzel from the Institute of Microtechnology, University of Hannover and S. Rudolph for discussions and providing us with the CMP slurry and polishing pad. This work was supported by the EU within the 6th framework program (Contact No. RII3-CT-2004-506008).

ACKNOWLEDGMENTS

V. CONCLUSIONS

We demonstrated for the first time that chemical-mechanical polishing of nickel zone plates with structures down to 25 nm width is possible. This is an important step toward the nanofabrication of zone plates with very high aspect ratios using the proposed stack process. In addition, such a process paves the way toward volume zone plates with sub-10 nm resolution. The demonstration of polishing nickel structures with line width in the 25 nm range by CMP is also an important result for the semiconductor industry since similar processes are required for the fabrication of future microprocessors with smaller interconnect linewidth. The thickness variation across the zone plate will be improved by applying pulse plating.

The polished zone plates have to be fabricated on a thick substrate which has sufficient mechanical stability for the CMP process. For using the zone plates as objectives in x-ray microscopes, the substrate has to be thinned. Backside thinning of the substrate can be done by wet chemical etching. However, a process to thin the substrate and protect the polished zone plate at the same time has to be developed.