Near-field stacking of zone plates in the x-ray range

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ABSTRACT

We use Fresnel zone plates as focusing optics in hard x-ray microprobes at energies typically between 6 and 30 keV. While a spatial resolution close to 0.1 µm can currently be achieved, highest spatial resolution is obtained only at reduced diffraction efficiency due to manufacturing limitations with respect to the aspect ratios of zone plates. To increase the effective thickness of zone plates, we are stacking several identical zone plates on-axis in close proximity. If the zone plates are aligned laterally to within better than an outermost zone width and longitudinally within the optical near-field, they form a single optical element of larger effective thickness and improved efficiency and reduced background from undiffracted radiation. This allows us both to use zone plates of moderate outermost zone width at energies of 30 keV and above, as well as to increase the efficiency of zone plates with small outermost zone widths particularly for the energy range of 6 – 15 keV.

1. INTRODUCTION

Fresnel zone plates are used as focusing optics with sub-micron resolution in hard x-ray microprobes at photon energies from a few keV to tens of keV.[1,2] Zone plates are circular diffraction gratings with radially increasing line density. They image an incident wave front into a series of real and virtual images in positive and negative diffraction orders. The performance of zone plates is typically characterized by (i) the resolution limit δ_m and (ii) the diffraction efficiency η_m in the m^{th} diffraction order.

(i) The resolution limit δ_m is directly related to the outermost zone width dr_n. A convenient measure for the resolution limit is the Rayleigh criterion for incoherent illumination, which yields the relation $\delta_m = 1.22 \cdot dr_n/m$ for the smallest features that can be obtained with a zone plate with outermost zone width dr_n. To improve the resolving power of zone plate in a particular diffraction order, the outermost zone width has to be reduced. Improving the resolution while maintaining reasonable diffraction efficiency is the major challenge in manufacturing zone plates for sub-100 nm imaging in the hard x-ray range.

(ii) The diffraction efficiency η_l of a zone plate is determined by the complex refractive index $\tilde{n} = 1-\delta - i\beta$, and geometrical parameters. such as the grating profile. The decrements δ and β are related to the phase shift and to photoelectric absorption of x-rays inside a medium, respectively.[3] The efficiency η_l at a given energy changes as a function of the thickness t of the zone plates.[4] At an optimized thickness $t_{opt,}$ the competing effects of phase shift and absorption are balanced, the efficiency for the diffracted wave is maximized, and the efficiency of the undiffracted order is minimized. By manufacturing zone plates with a thickness close to the optimized thickness, the focused flux and the signal/background ratio are maximized. Figure 1 illustrates the energy transfer between the undiffracted and the 1^{st}- and 3^{rd}- order diffracted wave for a gold zone plate with rectangular grating profile at a photon energy of 8 keV.

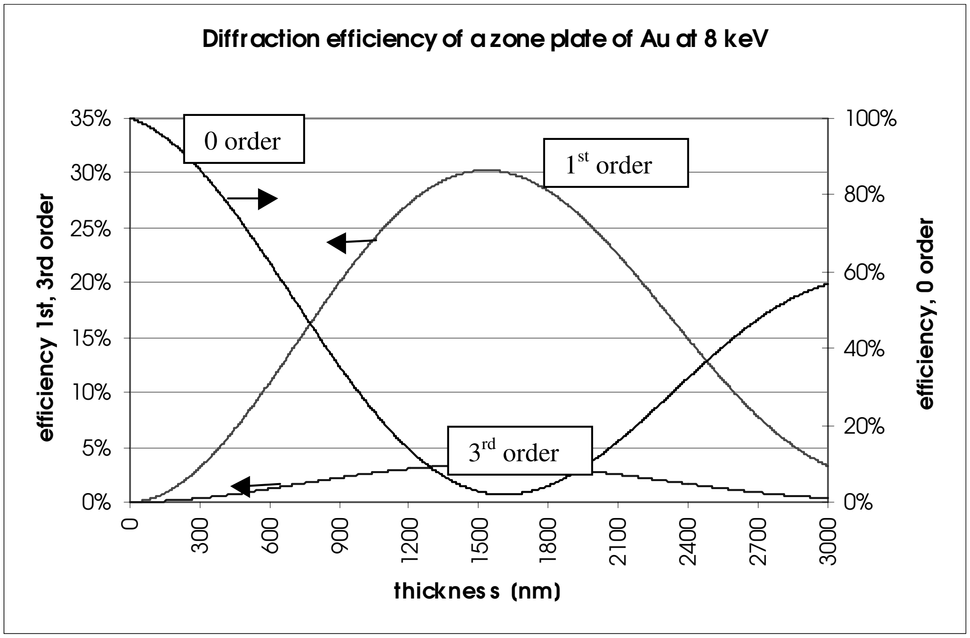

Fig. 1: Diffraction efficiency in the undiffracted and the +1st and +3rd diffraction order of a gold zone plate with rectangular profile for a photon energy of 8 keV. Efficiencies for the 1st and 3rd order are on the left axis, for the 0 order on the right. A maximum efficiency of 30% can be reached for an optimized thickness of 1.5 μm. For this thickness, the undiffracted order is reduced to 3.4%, thereby maximizing the signal/background ratio on the focal spot or image.

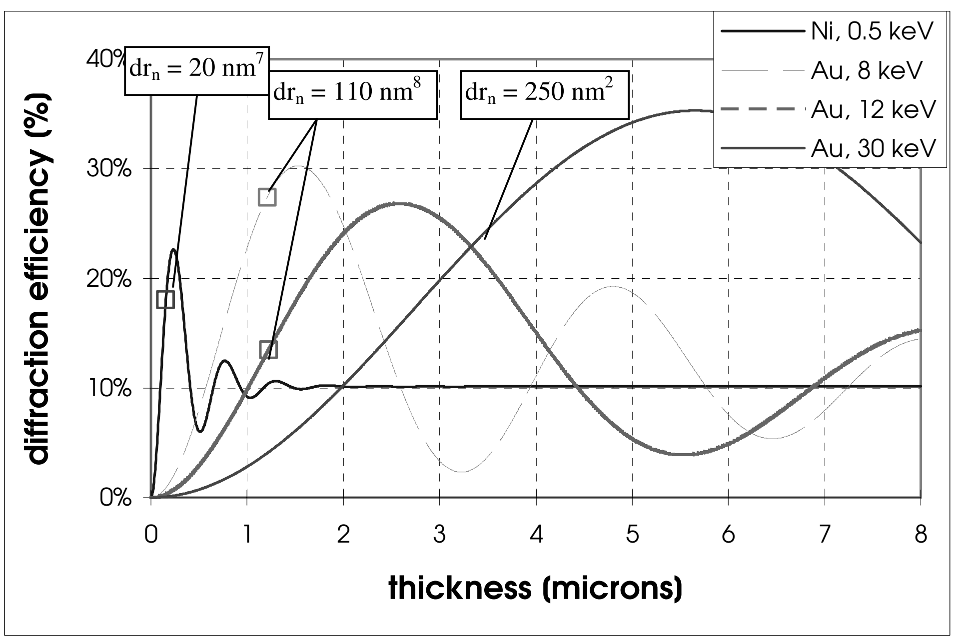

Fig.2: Diffraction efficiency vs. zone plate thickness for different x-ray energies. The curves were calculated based on ref. 4., assuming a rectangular grating profile. The calculation assumes that finite thickness effects do not have to be taken into account,[5] i.e., the zone width has to be reasonably large (on the order of 20 nm or larger for energies above 0.5 keV). The calculations have used the complex atomic scattering factor from Gullikson et al.[6] Outermost zone widths achieved in the different energy regions are marked in the graph. [7,8,2]

The two parameters that determine the performance of the zone plate, the outermost zone width dr_n and the thickness t, can be combined in a figure of merit, the aspect ratio $A:=t/ dr_n$. To achieve a Rayleigh resolution of 120 nm at a photon energy of 8 keV, a zone plate with an outermost zone width of 100 nm has to be manufactured. To obtain the maximum diffraction efficiency, a thickness of 1.5 µm, corresponding to an aspect ratio of 15 in the outermost zones, has to be achieved. For a zone plate with an outermost zone width of 20 nm, the required aspect ratio would be 75. Current fabrication techniques only allow manufacture of aspect ratios of around 10:1 for a small outermost zone width.[7,8] Therefore, highest resolution can be obtained only at a significant loss of efficiency and an increase in the signal/background ratio.

With increasing photon energy, the required thickness t_{opt} increases due to reduced interaction of x-rays with matter. For fixed outermost zone width dr_n, the aspect ratio therefore increases. Figure 2 compares the (first-order) diffraction efficiency for zone plates in different energy regions. A nickel zone plate used at an energy of 0.5 keV (soft x-ray region) requires a thickness of 230 nm to achieve a maximum diffraction efficiency of 23%. A gold zone plate used at 8 keV requires a thickness of 1.5 µm and a thickness of 5.7 µm at 30 keV. On the graph, typical values for the outermost zone width that have been obtained in the different energy ranges are marked. At an energy of 8 keV, the efficiency of a zone plate with an outermost zone with of 100 nm would be limited to 17% for an aspect ratio of 10:1 and to 7% for a photon energy of 12 keV. This results in significantly reduced focused flux in a hard x-ray microprobe, and, in the case of full-field-type transmission x-ray microscope, to increased radiation damage to the specimen. By stacking two or more zone plates on-axis, the thickness of the individual zone plates is added, and manufacture limitations in the fabrication of high aspect ratios be circumvented. [9] In the following, we will discuss the requirements for aligning several zone plates and show experimental results.

2. NEAR-FIELD STACKING OF ZONE PLATES

2.1 Near-field condition

If two zone plates are to perform as one optical element, they have to be positioned laterally and longitudinally within reasonable proximity. For the lateral alignment δ_t, a typical requirement is $\delta_t < 1/3\, dr_n$.[10] For longitudinal alignment, we require that both zone plates are aligned to a proximity within the optical near-field, $p \cdot \lambda < \pi/4d^2$, where d is the diameter of an aperture. This yields the condition for the proximity for a near-field stack of zone plates:

$$p < 0.76 \cdot dr_n^2/\lambda. \qquad (1)$$

The required proximity of the two zone plates corresponds to approx. 40% of the depth of focus of a zone plate, $\delta_l = 2 \cdot dr_n^2/\lambda$. Table 1 shows the required proximity of zone plates with outermost zone widths of (i) $dr_n = 20$ nm, the current state-of-the-art in the soft x-ray range,[7] (0.25 – 1 keV), (ii) $dr_n = 100$, typical for the 5-15 keV range,[8] and (iii) $dr_n = 250$ nm, as we have used at high energies (40-50 keV).[11] If the resolution requirement is relaxed at high energies, the required proximity is on the order of a millimeter or more, and three zone plates and more can be stacked. On the other hand, for achieving high spatial resolution below 30 nm in the multi-keV range, a proximity on the order of microns is required, and even stacking two zone plates in optical near-field will be challenging. For the current spatial resolution of 100-150 nm in the 6-15 keV range, two zone plates with a thickness of 0.8 - 1 µm can easily be stacked, yielding close to maximum efficiency and signal/noise ratio.

Table 1: Proximity condition for stack of zone plates vs. outermost zone width and photon energy. Note that, for an outermost zone width of $dr_n = 20$ nm at 8 keV, the near-field condition requires a proximity of 2 µm. For high-energy applications at reduced spatial resolution ($dr_n = 250$ nm), the proximity condition is rather relaxed (p = 1.5 mm at 40 keV), and stacking of three or more zone plates can easily be achieved.

dr_n [nm]	20		100		250	
Energy [keV]	8	40	8	40	40	100
p [µm]	2	10	51	253	1583	3957

We have performed dynamical calculations using coupled wave theory, and could verify that the zone plate efficiency is only little affected as long as the zone plates are in a proximity given by eq. (1).[12]

2.2 Experimental Setup

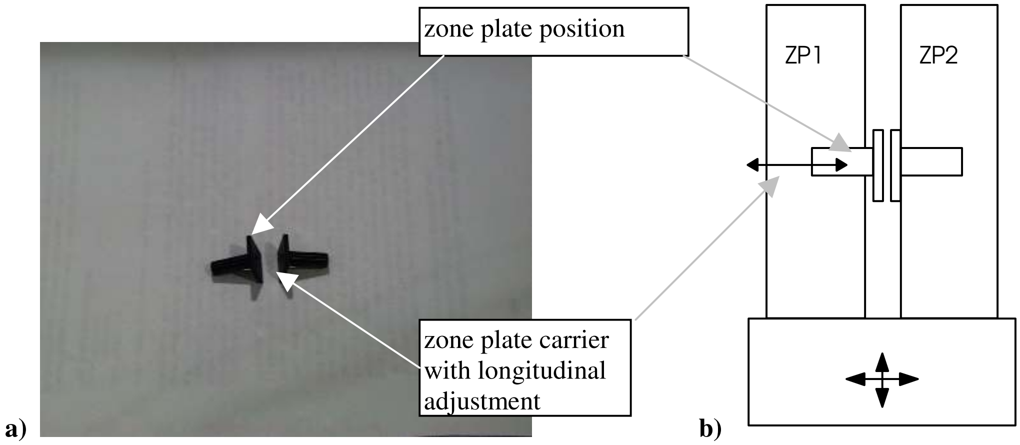

Fig. 3: Layout of a light-weight, stiff stage for alignment of two zone plates in close proximity. Each zone plate is positioned with a simple, commercial flexure stage (New Focus). Two of these flexure stages are mounted parallel to each other on an adapter plate (b). Guiding rails on the adapter plate prevent the flexure stages from rotating with respect to each other and keep the zone plates parallel. The zone plates are glued with nail polish onto a cylindrical carrier (a), which can be moved longitudinally within the flexure stage to minimize their proximity. One flexure is prepositioned using set screws, the second one can be aligned remotely using piezo-micrometer actuators (PMA). The adaptor plate with both zone plates, as well as a central stop, is mounted on an x/y stage for zone plate positioning in the hard x-ray microprobe.

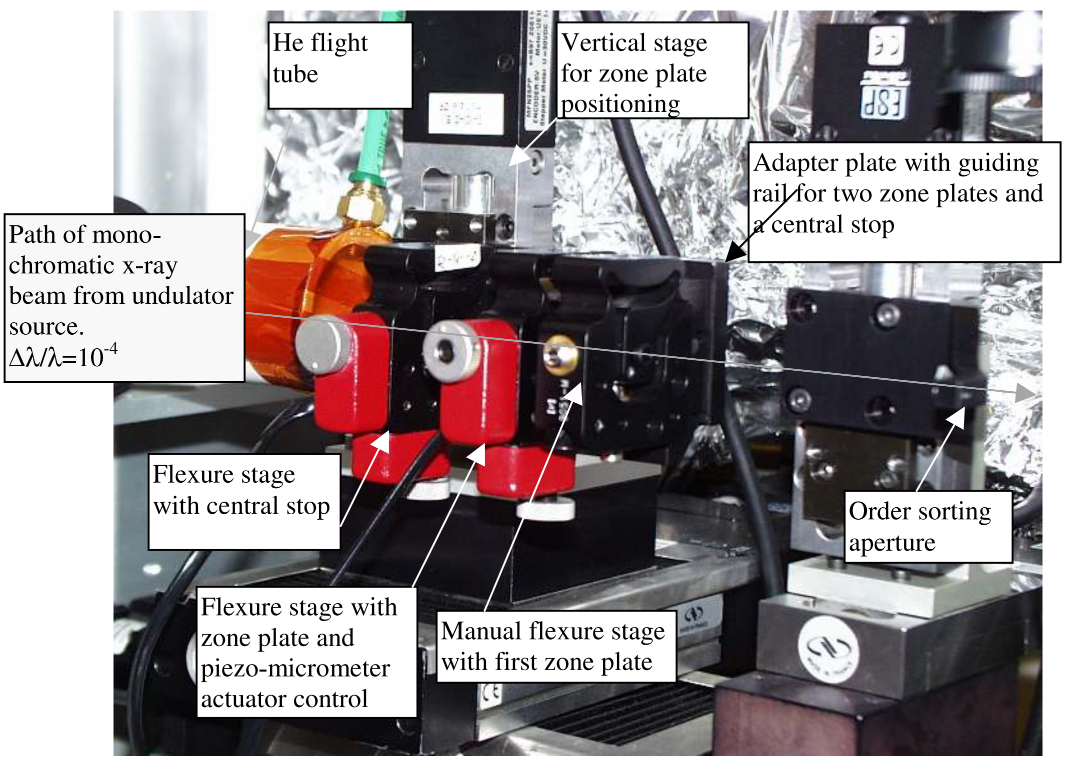

Fig. 4: Double zone plate holder with central stop, mounted in the 2-ID-E hard x-ray microprobe at the Advanced Photon Source. A tunable, monochromatic x-ray beam enters through a He flight tube from the left side. Three flexure stages on a common rail carry two identical zone plates and a central stop. One zone plate

is pre-aligned mechanically. The second zone plate and the central stop are aligned with respect to the first zone plate using piezo-micrometer actuators (PMA). The central stop prevents undiffracted x-rays from reaching the specimen. The focused beam is filtered through an order-sorting aperture to prevent unwanted diffraction orders from reaching the specimen. Real-time alignment of the two zone plates is achieved using x-ray interference (see fig. 5).

We have performed experiments with stacked zone plates both at high energies of 40 keV and 50 keV,[11] and at multi-keV energies of 7-15 keV. While the former experiments were aimed at evaluating the use of zone plates at significantly higher energies than typical, the latter were to (i) maximize the focused flux in state-of-the art hard x-ray microprobes and (ii) develop strategies for use in next-generation hard x-ray probes, which are designed to achieve a spatial resolution in the range of 30 nm and below.

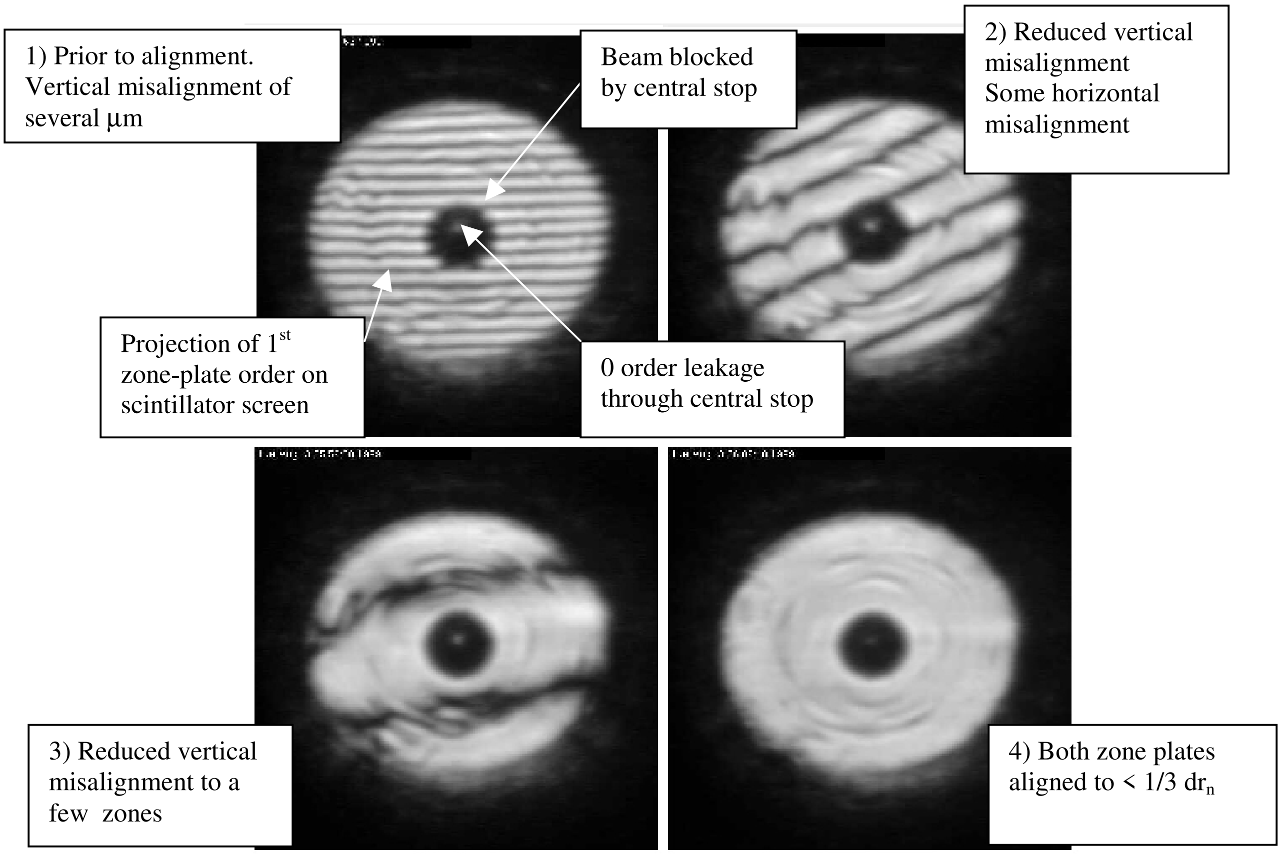

Fig. 5: Real-time alignment of two zone plates using their x-ray interference patterns. The interference pattern was recorded on a $CdWO_4$ scintillator crystal downstream of the zone plate pair, and the optical emissions from the crystal observed on a video camera with a 20x microscope lens. Alignment is achieved by driving the first zone plate horizontally and vertically. Images are recorded of the 1st zone plate order in transmission, seen from approx. 1 m downstream of zone plates through a 20 μm pinhole (order-sorting aperture). The interference patterns were recorded at the 2-ID-D hard x-ray microprobe.

For use at existing hard x-ray microprobes, we have designed a simple stage that allows the alignment of two zone plates with small outermost zone widths in very close proximity with high lateral precision and good stability (Fig. 3). Core piece of the stage are two commerical x/y flexure stages, one mechanically driven, the other one driven by piezo-micrometer actuators (PMA) (New Focus). The stages are placed on an adaptor plate, which maintains parallel alignment of the stages by using guiding rails. The zone plates are glued with nail polish on small aluminum carriers, which are inserted into the central hole in the flexure stage. The holders can be moved longitudinally to minimize the proximity between the two zone plates, and can be fixed in place using set screws. The adapter plate holds two flexure stages for zone plates and a third one for a central stop. The holder with the two zone plates and the central stop is mounted on a motorized x/y stage for alignment for the zone plate/central stop package with respect to the incident beam (Fig. 3). The setup also allows controlled displacement of two zone plates by a fraction of a zone width for differential interference contrast imaging.[13]

Alignment of a pair of zone plates is achieved in real time using x-ray interference. A $CdWO_4$ crystal is mounted approx. 1 m downstream of the microprobe and converts x-rays to visible light. The visible light emission is observed through a 20 x lens with a video camera. This system captures the x-ray intensity distribution downstream of x-ray optics and specimen. When both zone plates are aligned laterally to within several micrometers, their interference pattern can be resolved by this setup (Fig. 5). Using the PMA actuators, one of the zone plates is moved successively in x and y until no interference fringes are visible. The procedure takes approximately 2-3 minutes. The setup is stable to approx. 30 nm for several days, which is well matched to a typical experiment of 5-6 days.

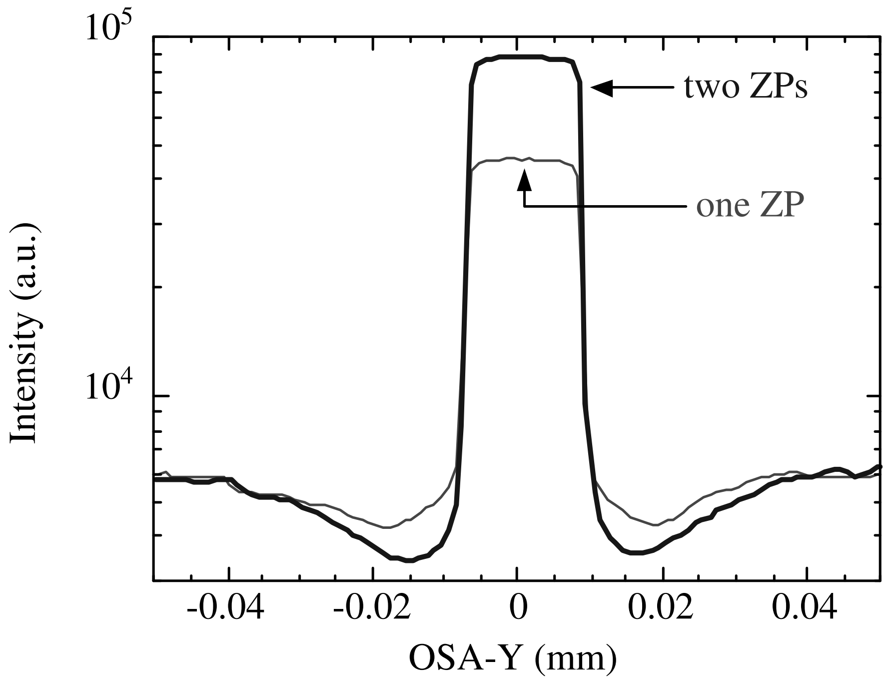

Fig. 6: Two zone plates with outermost zone width of dr_n = 100 nm and an outermost zone width of 0.8 µm aligned in optical near-field at an energy of 8 keV. A 20 µm pinhole was placed in the focal plane of the zone plate stack and scanned vertically through the focused beam. The graph shows measured counts in an ion chamber as function of pinhole position. A single zone plate provides a focused flux of approx $4.5 \cdot 10^4$ counts/s, corresponding to approx. 10^9 photons/sec, and a background of $5 \cdot 10^3$counts/s. Stacking two zone plates achieves a focused flux of $9 \cdot 10^4$ counts/s, corresponding to approx. $2 \cdot 10^9$ photons/sec, and a background of $3.5 \cdot 10^3$ counts/s. The x-ray flux is plotted on a logarithmic scale to show variation of the background.

Figure 6 shows the increase of focused flux and the reduction of background from the undiffracted x-ray beam that reaches the specimen through the order-sorting aperture. We used two identical zone plates with a thickness of 0.8 µm at a photon energy of 8 keV and a bandwidth of $\Delta E/E = 10^{-4}$. A central stop with 30 µm diameter is placed upstream of the zone plate stack to block undiffracted beam and higher diffraction orders. A 20 µm pinhole is scanned vertically through the focal plane. The transmitted photon flux is recorded using an ion chamber, and converted to a count rate using a charge-sensitive preamplifier and a voltage-to-frequency converter. A count rate count of $4.5 \cdot 10^4$ counts/s correspond to a flux of approx. 10^9 photons/sec at 8 keV.

With a single zone plate, a count rate of $4.5 \cdot 10^4$ counts/sec is measured in the focus, and a background of $5 \cdot 10^3$counts/s outside of the focus. Two well-aligned zone plates yield a count rate of $9 \cdot 10^4$ counts/s in the focus and a background of $3.5 \cdot 10^3$corresponding to a gain of 2 x in focused flux and of 3.5 x in signal/background.

CONCLUSIONS

Near-field stacking of zone plates is a strategy that effectively doubles or triples the aspect ratio that can be achieved with a single zone plates, while maintaining the outermost zone width and, thereby, the spatial resolution. Near-field stacking allows a significant increase in the diffraction efficiency and improvement in the signal/background ratio in the focus without having to increase the aspect ratio during zone plate manufacture. Since achieving the high aspect ratios required in the multi-keV and hard x-ray range is limited by pattern transfer techniques, this approach will be critical in achieving a spatial resolution of 30 nm or less in hard x-ray microprobes. It can also be used to modulate the grating profile, e.g., to achieve blazing by stacking zone plates with different line-space ratios to approach a triangular zone profile. A simple stage that accommodates two identical zone plates has been in use in the hard x-ray microprobes at sector 2 of the APS for several years. Alignment is achieved within minutes using x-ray interference; the stack is stable in the experimental setting for several days. The setup is designed for easy lateral alignment using real-time x-ray interference patterns, and is therefore well suited for differential interference contrast imaging.

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