We demonstrate the fabrication of a 2D Compound Array Refractive Lens (CARL) for multi-contrast X-ray imaging. The CARL consists of six stacked polyimide foils with each displaying a 2D array of lenses with a 65 μm pitch aiming for a sensitivity on sub-micrometer structures with a (few-)micrometer resolution in sensing through phase and scattering contrast at multiple keV. The parabolic lenses are formed by indents in the foils by a paraboloid needle. The ability for fast single-exposure multi-contrast imaging is demonstrated by filming the kinetics of pulsed laser ablation in liquid. The three contrast channels: absorption, differential phase and scattering are imaged with a time resolution of 25 μs. By changing the sample-detector distance it is possible to distinguish between nanoparticles and microbubbles, respectively.

Keywords: laser ablation in liquids, multi contrast retrieval, scattering contrast imaging, X-ray lens.
FIG. 1. (a) Scheme of the fabrication of the compound array refractive lens by first a sequential embossing of lenses in foils and later successive stacking of these foils and fixation. The diagonal distortion is linked to the pyramidal foil bending. (b) Image of a single foil displaying the uniformity of the array. The elevation of the pyramid is estimated to be less than 400 μm. Thus, the plastic strain is symmetrically shifted towards the outer array parts. Due to this plastic flow in the embossing process a slight pyramidal shape develops in the lens array (visible in Fig. 1 (b) as a slight defocusing of the center in the optical micrograph and a hint of pyramidal facets). The array size of successive foils in the final stack is enlarged by one line of lenses each, starting with an array of 99x99 lenses. This aids the alignment of the semi-transparent foils under the microscope. Starting with the largest lens array the next-in-line array is positioned precisely on top by a manual goniometer and glued in place (see Fig. 1 (a)). A slight (<5 μm) misalignment of lenses in a CRL is mainly changing the absolute transmission of the CRL, which is uncritical in our case.

The characterization of the CARL and multi-contrast imaging of the ablation process were performed at the synchrotron at KIT (Karlsruhe, Germany), at the tomography instrument TOPO-TOMO. For characterization and PLAL imaging monochromatic X-rays at 9 keV (bandwidth 2%) and a white beam (central energy 15 keV, filtered by 0.2 mm of Al) were used, respectively. X-rays were detected by a CMOS camera (Andor Neo with 10 μm LSO:TB and PCO.dimax with 50 μm LuAg:Ce scintillator for focus measurement and PLAL, respectively). Corrections for dark current and flat-field distortions were performed (illumination without X-rays and 10 images before laser action, respectively). This setup is illustrated in Fig. 2. The X-rays coming from the synchrotron were concentrated into the beamlets by the CARL (see Fig. 1 (c)). The sample was placed between the CARL and the detector. Technically, it is also possible to place the sample in front of the CARL as usually done in GI. Here we need a short distance between the sample and the detector for setting the length scale in scattering contrast. The CARL was placed at a distance to the detector, which corresponded to the calculated focal length for 15 keV. Note that the point of tightest focus was slightly broadened due to the usage of a filtered white beam. Time resolution was gained by operating the active-pixel camera (PCO.dimax) with a frame rate of 10 kHz and 30 μs exposure per frame. A fourfold interleaving of records with shifted time delay between laser and camera results in an effective frame rate of 40 kHz. A delay generator (Research Instruments, DG535) controlled the delay between laser and camera.

The ablation process was performed in a 3D-printed flow chamber. The chamber design and functionality was described elsewhere. In brief, the chamber had a rectangular reaction volume of 0.5 ml with channels providing an optimized water flow. The laser was focused onto the target by a lens, which also acted as a chamber seal. The side walls were sealed by Kapton foils, allowing for X-ray transmission. As target a Zn wire (1 mm, Advent, 99.99%) was used and continuously transported (perpendicular to laser and X-ray beam) to obtain a clean surface spot for each individual laser shot. The wire was suspended in water being clamped on each sides 5 mm away from the ablation spot. The laser was a Nd:YAG laser (wavelength 1064 nm, Continuum Minilite I) with 10 mJ pulse energy. An average over 500 shots was acquired for each distance, taking advantage of the repeatability of the process.

The general image and contrast formation have been described in earlier publications. The overall performance of the different optical elements depends on the beam structuring (visibility) and on the retrieval algorithm. The CARL creates a spot for each CRL stack on the detector. The change of each of these spots by the sample in intensity, position and width corresponds to absorption, differential phase and scattering, respectively. It should be noted that the distinction between phase and scattering contrast is interdependent of each other and reflects on the geometry and size of the objects in the experimental setup. Also, crosstalk from absorption to scattering contrast is of importance for a proper data analysis.

We used 2D-Gaussian fit for the estimation of the...
FIG. 2. (a) Setup: X-rays from the synchrotron are focused into an array of beamlets by the CARL before intersecting the sample and being detected by a (fast) X-ray image detector. From the relative changes of intensity, position and width the different contrasts are reconstructed. Two sets of images at the large distance: (b)-(d) for the first cavitation bubble and (e)-(f) for the first rebound. (b) and (e) show the transmission (higher transmission = brighter), (c) and (f) the scattering contrast (higher scattering = brighter) and (d) and (g) the lateral differential phase contrast.

height, position and width of the spots (for more details see supplementary material, section I A) for the CARL characterization. It is also possible to retrieve absorption, differential phase and scattering contrasts from Fourier analysis, assuming that the spots reside on a quadratic array with equal spacing \( R = N \delta \) where \( R \), \( N \) and \( \delta \) are the radius of curvature at the apex, number of stacked lenses and the decrement of the index of refraction of the lens material. With an approximate radius of curvature of \( (25 \pm 5) \) \( \mu \)m and \( \delta = 3.76 \cdot 10^{-6} \) for polyimide at 9 keV, the expected focal distance is \( f_{\text{theo}} = (110 \pm 20) \) cm.

As shown in Fig. 3 (a) the position of minimal spot size (for a region of 20x20 spots, calculated by polynomial fit of order 2) is at 76 and 96 cm for x- and y-direction, respectively. This is in fair agreement with the expected value. The beamlet intensity also peaks at 78 cm. The slight difference in spot size in the two directions originates from the imaging of the synchrotron source size by the lenses. Placed at a bending magnet of the ring, the horizontal source size (with 2 mm primary slit opening) is larger than in vertical direction (0.2 mm). The finite source size may also be the reason for the rather flat distribution of focal spot size versus distance and the apparently decreased focus length. The geometric focus competes with the demagnification of source size at a relatively shorter distance.

Fig. 3 (c) and (d) display the spot width in x- and y-direction of 62x53 spots at a distance of 85 cm, with sufficient uniformity across the lens array. Again, the pyra-
agglomeration are still under investigation. Details of the interaction between bubble and NPs, such as redeposition and agglomeration, are still under investigation. Between bubble and NPs, such as redeposition and agglomeration, are still under investigation. Typical X-ray radiographs (transmission contrast) are displayed in Fig. 4 as inset (see supplementary material for a video with all contrasts). The fully extended (hemispherical) first bubble as well as the detached first rebound structure are found at delays of 100 µs and 325 µs, respectively. The latter is of complicated nature, as it does not seem to be a homogeneous cavity. Earlier SAXS measurements have shown mostly two size levels of produced NPs, one with diameters of approximately 10 nm and one with diameters larger than 40 nm in silver or gold. Zinc ablation produces larger particles (between 10 and 70 nm). Scattering contrast shows up where a sufficient concentration of structures within the sensitivity interval are located.

The peak sensitivity for both sample-detector distances changes from 100 nm for the short to 950 nm for the large distance. The normalized sensitivities are shown as inset in Fig. 4. It should be mentioned that the absolute sensitivity for the short distance is lower than that for the large distance. As consequence, smaller structures remain visible for an increase in detector distance, as seen earlier. Large structures only appear with the larger distance.

A clear difference in the signals for the short and large sample-detector distance is observed. Within the first bubble (0-250 µs delay) the signal of the short distance is higher compared to the first rebound (250-400 µs). This is in contrast to the signal evolution at the large sample-detector distance. Here, the signal within the first rebound is boosted considerably. Both signals return to almost 0 after the bubbles have vanished. The difference in size sensitivity allows identifying the signal at the short distance of being related to the ablated NPs within the bubble, while at the large distance the sensitivity on larger structures favors the notion of emerging microparticles. Additionally, agglomerated NPs may add to the signal at large distance. This is in line with findings in optical stroboscopy and X-ray radiography, where the first rebound and in particular the bubble stem were optically opaque and seemingly not homogeneous, showing micrometer scale metastable permanent gas bubbles.

The multi-contrast and in particular scattering imaging allows for the identification of nanoscale features during X-ray imaging. While the integral scattering signal does not allow resolving different sizes directly, a variation of sample-detector distance can coarsely discriminate different size fractions.

We demonstrated a facile route for fabricating a 2D array of X-ray lenses by sequential stamping into a polymer foil. We realized an area of 99x99 lenses and 6.5x6.5 mm. Lateral size is further scalable, the pattern is easily changeable and the focal distance can be changed by the needle shape or further stacking. This CARL allows for the simultaneous assessment of absorption, differential phase and scattering in single-exposure X-ray measurements. By using a CARL the local flux density is increased compared to the use of Hartmann masks leading to decreased exposure times. The increased flux density...
By using the differential phase the CARL can also be used as a Shack-Hartmann sensor for hard X-rays (SHARX)\textsuperscript{7}. Further improvements in the fabrication can be obtained by stamping with needles of better defined shape and needle arrays. A central parameter to improve is the visibility contrast which can be achieved by higher fill factors in the lens plane. To improve the scattering sensitivity further one could also produce different pitches in x- and y-direction to gain results of different sensitivities within one measurement\textsuperscript{41}.

See supplementary material for (i) the detailed description of the multi-contrast retrieval, (ii) multi-contrast images of the first rebound and (iii) a video of the important contrasts of the ablation process.

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(a) Synchrotron hard X-rays are directed at a target. A laser is fired, causing a bubble to form and rebound. The resulting images are captured by a detector.

(b) Initial image with a bubble of diameter 1 mm.

(c) Follow-up image showing the bubble's rebound.

(d) Another follow-up image.

(e) High-resolution image showing detailed structure.

(f) Additional high-resolution image.

(g) Further high-resolution image.