Arresting sample explosion for X-ray imaging

S.P. Hau-Riege[1], S. Boutet[2], A. Barty[1], S. Bajt[3], M. Bogan[2], J. Andreasson[4], B. Iwan[4], M.M. Seibert[4], J. Hajdu[4], A. Sakdinawat[5], J. Schulz[6], R. Treusch[3], and H.N. Chapman[3,7]

[1] *Lawrence Livermore National Laboratory, 7000 East Avenue, Livermore, CA, 94550*

[2] *SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, California 94025, USA.*

[3] *HASYLAB, Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany*

[4] *Laboratory of Molecular Biophysics, Department of Cell and Molecular Biology, Uppsala University, Husargatan 3, Box 596, SE-75124 Uppsala, Sweden*

[5] *University of California, Berkeley, 253 Cory Hall, Berkeley, CA 94720*

[6] *Centre for Free Electron Laser Science, DESY, Notkestraße 85, 22607 Hamburg, Germany*

[7] *Universität Hamburg, Notkestraße 85, 22607 Hamburg, Germany*

Extremely intense and ultra-short x-ray pulses from free-electron lasers (FELs) open up the possibility for high-resolution structural studies on atomic clusters, macromolecular complexes, or larger structures without the need of crystallization [1]. Femtosecond pump-probe experiments also enable the exploration of dynamics processes, including chemical reactions. Such experiments require high photon fluences (~10^{12} photons/molecule) and ultra-short X-ray pulse lengths. Predictions suggest these requirements can be significantly relaxed, if the sample is surrounded by a sacrificial tamper layer [2, 3] to dampen the Coulomb explosion by providing a bath of photo-induced free electrons, and arresting the hydrodynamic expansion of the sample through inertial confinement.

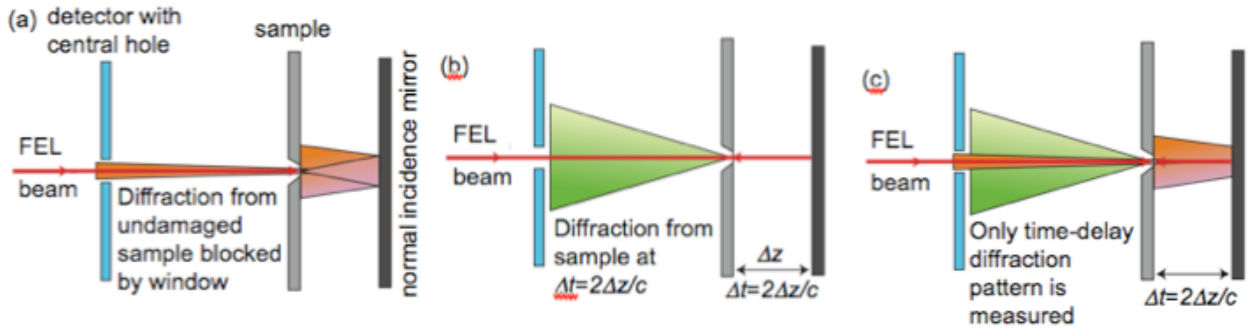

Figure 1: Scattering geometry of the one-color pump-probe experiments. (a) The FEL beam irradiates the sample, initiating the reaction, and reflects from a normal-incidence multilayer mirror back onto the sample [5]. (b) The FEL beam once again illuminates the sample with a time delay of $\Delta t = 2\,\Delta z / c$, probing the reaction. (c) The prompt diffraction signal is blocked using a small window sample. In this way only diffracted light from the time-delayed sample is detected using a charge-coupled device (CCD) on the left.

Here we describe results from ultra-fast time-resolved diffraction experiments with coherent 0.1 keV X-rays from the FLASH soft x-ray FEL [4], showing that the sample lifetime can be extended to several picoseconds under these conditions. Figure 1 shows the experimental arrangement used here. By reflecting the FEL beam back onto itself the same beam can be used both to trigger the sample explosion and then to probe the exploding sample with a well-defined time delay. In the variation used here, we placed samples on very small sample windows, which prevent the primary diffracted light from reaching the detector after it is reflected back from the mirror.

The samples for measuring the hydrodynamic explosion rate consisted of cylindrical aluminum pillars deposited on 100 nm-thick silicon-nitride membranes, spanning 7.5 µm square windows etched into a silicon wafer (see inset in Figure 2c). Far-field diffraction patterns measured with and without a tamper for time delays of 25 fs and 13.5 ps are shown in Figures 2 (a) and (b). A circular pillar gives rise to a diffraction pattern similar to Airy's rings with the angular positions of intensity minima inversely proportional to the pillar radius. In our experiment this single-pillar pattern is

modulated by speckles that encode the arrangement of the identical pillars. The expansion of the pillars causes a change in the Airy-like envelope of the diffraction pattern. The diffraction patterns before the explosion are averages over multiple low-fluence exposures during which the sample did not change significantly (the "unperturbed" sample) .We use these patterns as a basis for comparison. The diffraction patterns with 13.5 ps time delay were obtained from single-pulse high-fluence exposures. The difference between tampered and untampered sample is immediately apparent. With a tamper layer present, the diffraction patterns of the low- and high-fluence exposures are similar and the diffraction Airy-like minima are at similar positions. Without a tamper layer, the diffraction minima have shifted to significantly smaller angles, indicating that the aluminum pillars have expanded. Reconstruction of the diffraction patterns using iterative phase retrieval enables us to directly image the particle expansion. Figures 3 (c) shows the difference of reconstructed images before and during the explosion, both with a tamper of the aluminum pillars and without, giving even more detailed information about the expansion process. Note that our results also indicate that tampering could produce atomic resolution in "diffraction before destruction" experiments at hard x-ray FELs, which will enable groundbreaking new capabilities in various disciplines, including structural biology, cluster physics, and nanoscale chemistry.

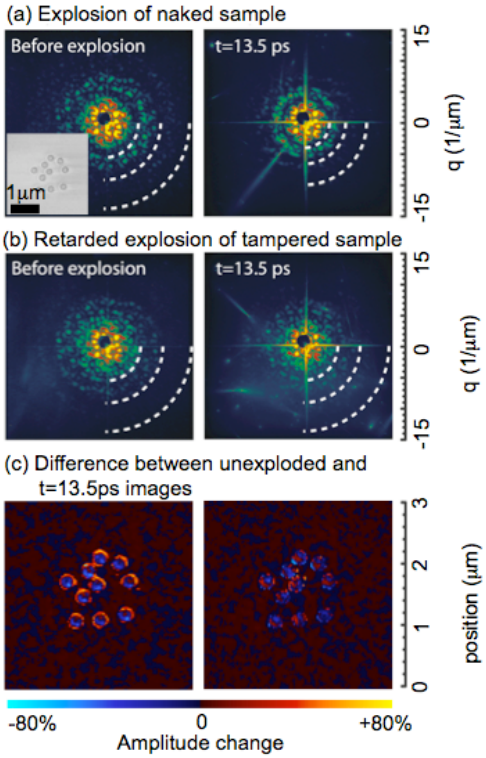

Figure 2: Measured far-field diffraction patterns and reconstructed structures. (a) Diffraction from the naked, untampered sample (with an SEM picture on the left of the sample before exposure), and (b) from the tampered sample. Arcs highlight Airy-like diffraction minima in the measured data. Contraction of the arcs indicates sample expansion. This is significant in (a) and small in (b). (c) Difference images of reconstructed structures. Light colors indicate significant density changes. The untampered sample (left) shows large expansion, whereas the tampered sample (right) shows minimal structural changes. The wavelength was 13.5 nm, the pulse length 25 fs, and the fluence 31 J/cm^2.

References

[1] R. Neutze,W. Wouts, D. van der Spoel, E. Weckert, and J. Hadju, Nature **406**, 752 (2000).

[2] S.P. Hau-Riege, R.A. London, H.N. Chapman, A. Szoke, and N. Timneanu, Phys. Rev. Lett. **98**, 198302 (2007).

[3] A. Mikaberidze, U. Saalmann, and J. M. Rost. Phys. Rev. A **77**, 041201 (2008).

[4] W. Ackermann et al., Nature Photon. **1**, 336 (2007).

[5] H.N. Chapman et al., Nature **448**, 676 (2007).