Lensless MAD Imaging of Nonperiodic Nanostructures

The intricacy of structure determination due to the lost phase information in diffraction measurements can be downsized to a manageable problem by labeling the unknown specimen with a few heavy atoms that serve as reference scatterers. In this way, multiple-wavelength anomalous diffraction (MAD) phasing has revolutionized macromolecular structure determination on atomic length scales and has become a well established technique in x-ray crystallography with dedicated synchrotron beamlines.[1] An important prerequisite for conventional MAD is the periodicity of the sample which requires for assembling the biological molecules into a crystal.

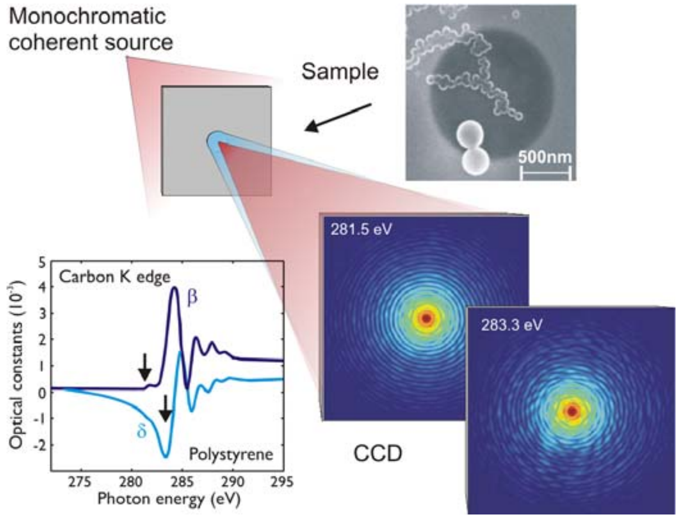

Figure 1: MAD imaging setup: The sample (SEM image) is illuminated with a monochromatized and spatially coherent source (red). MAD phasing exploits the energy-dependent interference of the resonant exit wave (red) with the nonresonant exit wave (blue). The interference patterns recorded with a CCD detector reveal notable changes in vicinity of the carbon *K* edge. The exposure times were in the range of 700-1000 seconds with a coherent flux of 10^6 photons s^{-1} μm^{-2}. The corresponding optical constants for the selected energies are indicated in the plot.

In this work, the methodology of MAD is extended to non-periodic structures using the concept of coherent x-ray imaging, cf. Figure 1. The solution of the phase problem is demonstrated in a proof of principle experiment by a combination of two resonantly recorded scattering or speckle patterns at the carbon *K* edge. This new approach merges iterative phase retrieval [2] and x-ray holography approaches [3] and facilitates unique and rapid reconstructions. The inherent resonant aspect provides sensitivity to the elemental, chemical and magnetic state that further renders lensless MAD imaging widely applicable to a broad range of nanostructures with in principle wavelength limited spatial resolution.

The experiment was carried out on the soft x-ray coherent scattering Beamline 13-3 of SSRL. The speckle patterns were recorded from dispersed polysterene spheres on a silicon nitride membrane with a gold aperture. The scattered wave field consists out of resonant and nonresonant components and the interference of both is recorded with an array detector in the far-field of the sample. The strong change of the optical constants or atomic scattering factors of matter across resonances at the threshold of core electron excitation energies allows for defining a generalized reference wave as the energy-dependent part which resonantly phases the nonresonant scattered wave emerging from other parts of the sample.

The interference patterns reveal remarkable changes at nearly the same photon energy reflecting the changes in the optical constants. The optical constants have been determined by measuring the absorption of the sample. Given the optical constants and applying the MAD concept the energy-dependent interference patterns have been decomposed into resonant and non-resonant amplitudes and their relative phases (MAD phases). The non-

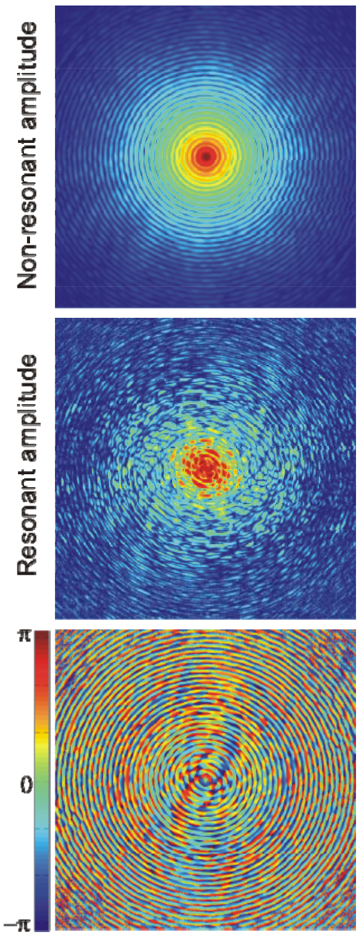

Figure 2: MAD phasing results are used as reciprocal space constraints to reconstruct the image of the specimen: (top) the non-resonant amplitudes and (center) the resonant amplitudes of the scattered waves. Intensities are shown on the same logarithmic scale. (bottom) The MAD phases represent the phase relation between the two scattered waves. 800x800 pixels are shown corresponding to a momentum transfer of 0.153 nm^{-1}.

resonant amplitude reproduces an Airy pattern from the circular Au aperture while the resonant part shows a speckle pattern from the local arrangement of the polysterene spheres.

Two images, a resonant image of the polysterene spheres and a nonresonant image of the aperture, are simultaneously reconstructed by disentangling the MAD phases into resonant and nonresonant components. This is accomplished by applying the reference-guided phase retrieval (RPR) method which has been developed in this work. The MAD phases contain all information about the orientation of the sample removing arbitrariness in the reconstruction process shown in figure 3. After two iterations, the sample structure becomes apparent, consolidates over a couple of iterations, and refines in less than 30 iterations. The resolution of MAD imaging is ultimately given by the observation of interference terms in the speckle patterns at high momentum transfer. In the presented case the resolution of 25nm is limited by statistics.

Lensless MAD imaging can be viewed as an in-line x-ray holography technique which eliminates the need for fabricating small reference structures as in x-ray Fourier Transform Holography. In combination with the RPR algorithm the method is capable of providing unique and rapid reconstructions of a variety of inhomogeneous systems, representing regions with different atomic constituents, chemical composition such as organic functional groups, and magnetic behavior such as changes in moments and their orientations.

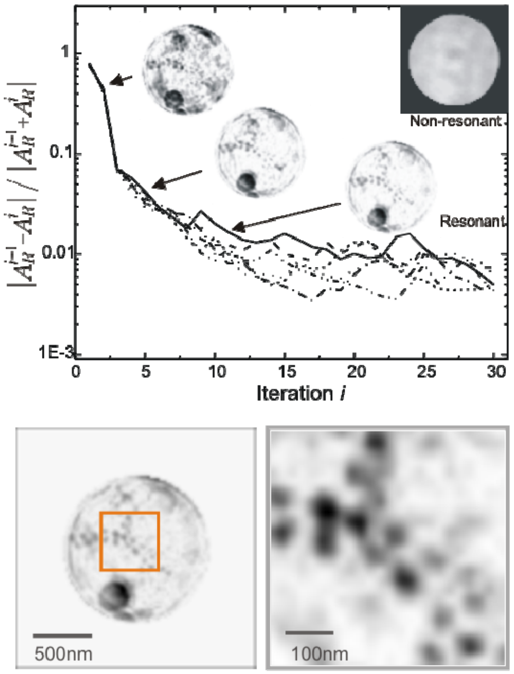

Figure 3: (top) The normalized convergence error is plotted against the iteration. The group of convergence curves represents different reconstructions, based on starting conditions with changes in the optical constants by 20%. The respective reconstructions are similar within 4% indicating the robustness of the RPR against uncertainties in the determination of optical constants. The insets show the progressing resonant structure determination and the nonresonant circular aperture (its simultaneous progression is not shown). (bottom) The final resonant reconstruction of the polysterene spheres at 25nm resolution and a magnified portion with adjusted linear contrast.

Primary Citation

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References

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[2] J. R. Fienup, J*. Opt. Soc. Am.* A 4, 118 (1987).
[3] S. Eisebitt *et. al.*, *Nature* (London) 432, 885 (2004).

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