acutely sensitive to local site symmetry. Furthermore, these inelastic images illustrate the complex nature of the scattering from the boron octahedra. The implications for extracting information about individual atoms within a specimen using conventional quantitative STEM and using direct images of the scattered electron probe will be discussed.

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Keywords: scanning transmission electron microscopy, electron scattering, double-aberration correction.

MS.44.5

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***QFocus*: Structure reconstruction from focal series of hrtem images**

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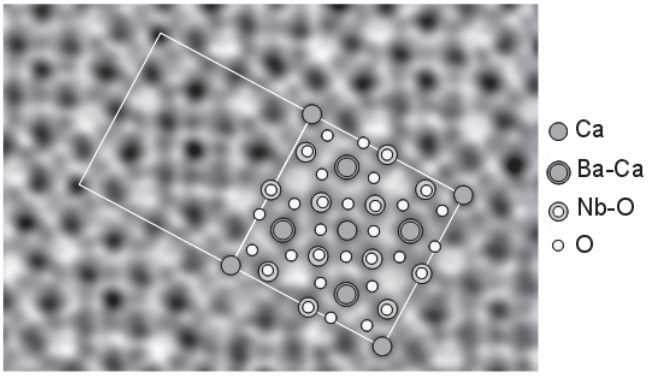
High-resolution transmission electron microscopy (HRTEM) images are usually not directly interpretable in terms of crystal structure due to various aberrations in the microscope lenses. Although significant progress has been made recently in the development of hardware aberration correctors, especially spherical-aberration correctors [1], the majority of transmission electron microscopes in use today are traditional ones and structure reconstruction by software methods still serves as a cost-effective alternative. Representative software methods include exit wave reconstruction using through-focus image series [2] and contrast transfer function (CTF) correction methods using single images or image series [3]. Here we present a CTF-correction based structure reconstruction method, using through-focus series of HRTEM images taken with a fixed focus step, in which all images in the series are corrected for CTFs and combined into a structure image. To determine the starting defocus of the series, trial defocus values in a large range are tested and CTF corrections are made to all images. The similarity of the phases of the Fourier transform between the corrected images is used as the criterion to judge whether a trial defocus value is close to the true one. The two-fold astigmatism is determined by dividing Fourier transforms into sectors and determining the defocus along different directions. As crystallographic phases are used in defocus and astigmatism determination, no amorphous areas are required in the images. On the other hand, since CTF correction is done for all pixels within the resolution limit in reciprocal space, this method works for both perfect and defect crystals.

The method was applied on a 20-image focal series of $Ca_{0.28}Ba_{0.72}Nb_2O_6$ (*P*4*bm*, *a*=12.43Å, *c*=3.96Å). The experimental images were collected with a focus step of -26.6 Å on a JEOL-2100F transmission electron microscope equipped with field emission gun. The starting defocus of the series is determined to be -260 Å and the two-fold astigmatism is 33 Å with an azimuth angle of 13° clockwise with respect to the *x*-axis of the images. The reconstructed image is shown in the figure below, in which atoms appear as black dots. Not only metals but also atomic columns as light as oxygen can be seen.

The present method has the advantages of automatic defocus and two-fold astigmatism determination without amorphous areas, less CTF crossover problems and improved signal-to-noise ratio as compared to CTF correction methods using single images. The method has been implemented in a user-friendly program, *QFocus*, which we hope may help non-TEM experts to use HRTEM images for solving their structure problems.



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Keywords: HRTEM, structure reconstruction, electron crystallography

MS.45.1

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"Making the molecular movie": First frames…coming features

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One of the great dream experiments in Science is to watch atomic motions as they occur during structural changes. In the fields of physics, chemistry and biology, this prospect provides a direct observation of the very essence of chemistry and the central unifying concept of transition states in structural transitions. From a physics perspective, this capability would enable the observation of rarified states of matter at an atomic level of inspection, with similar important consequences for understanding nonequilibrium dynamics and collective phenomena. This experiment has been referred to as "making the molecular movie". Due to the extraordinary requirements for simultaneous spatial and temporal resolution, it was thought to be an impossible quest and discussed in the context of the purest form of a gedanken experiment. Recent developments in femtosecond electron guns with sufficient brightness to even execute single-shot structural determinations have made this experiment become a reality [1]. Previously thought intractable problems in attaining sufficient brightness and spatial resolution, with respect to the inherent electron-electron repulsion or space charge broadening, have been solved. With this new level of acuity in observing structural dynamics, there have been many surprises and this will be an underlying theme. Several movies depicting atomic motions during passage through structural transitions relevant to condensed phase dynamics will be shown [2], [3], [4]. The primitive origin of molecular cooperativity has also been discovered in recent studies of molecular crystals. These new developments will be discussed in the context of developing the necessary technology to directly observe the structure-function correlation in biomolecules –the fundamental molecular basis of biological systems. The future is even brighter with the advent of a new concept in relativistic electron guns that will open up direct observation of atomic motions in solution and

gas phase systems with femtosecond temporal resolution to watch even the fastest atomic motions. Some of the important scientific problems to be addressed with ultrabright electron sources will be discussed to give an impression of the potential impact of this emerging field.

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Keywords: femtosecond electron diffraction, ultrafast structural dynamics, electron sources

MS.45.2

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Measuring femtosecond structural dynamics at a hard X-ray laser: challenges and successes

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The World's first hard X-ray laser, the Linac Coherent Light Source (LCLS), is now operational at the SLAC National Accelerator Laboratory [1]. The LCLS routinely produces femtosecond pulses of 9 keV photons with 2 mJ of pulse energy at a 120 Hz repetition rate. The unprecedented combination of photon flux, spatial and time resolution of the LCLS promises to revolutionize the observation of structural dynamics by measuring the time evolution of the electron density during a photo-induced transformation.

Although many of the attributes of X-ray laser are ideal for studying structural dynamics, there are significant experimental challenges involved that must be overcome before successful experiments are realized. Unlike storage ring based sources, X-ray pulses from the LCLS exhibit inherent fluctuations due to the self-amplified spontaneous emission (SASE) process through which the radiation is generated. As such the spatial, spectral, temporal and intensity properties vary on a pulse-by-pulse basis. These fluctuations are particularly problematic for experiments studying stimulated dynamics.

The first hard X-ray experimental station, the X-ray Pump-Probe (XPP) instrument, began user operations in the Fall of 2010. The XPP instrument is designed specifically for time-resolved studies and includes a synchronized optical laser. The first experiments included time resolved studies of optically initiated ferroelectricity, photoactive proteins (myoglobin) and iron spin crossover complexes. Many challenges associated with a SASE based X-ray source were encountered. However, most challenges were overcome and a time resolution of 150 fs was achieved.

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Keywords: instrumentation, laser

MS.45.3

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Structural dynamics of proteins conformers and conformer selection in chemical reactions

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Characteristic for all chemical reactions are bond breaking and bond making processes. Our vision is to optimize chemical reactions towards specific product states by a clever combination of chemical site-specificity, self-assembly and state-selectivity which can be "tuned" from orbital control through the structure of the local environment and selective excitation schemes (heat / optical pulses) to bulk structural changes – or to say it in other words – from the simple to the complex. We would like to understand - what are the driving forces of environment tuning chemistry? Most motors are defined through gradients in the chemical potential. Are chemical redox potential changes or the changes of chemical potential the most efficient chemical ways for storing energy?

To do so we need to gain a deeper understanding of the mechanism of chemical reactions from a structural point of view – besides its understanding of energetic. In order to elucidate information about reactions and their pathways multidimensional reaction landscapes are required for their description – not only in the energy coordinate but also in the reaction coordinate. In order to elucidate information about the reaction coordinate of complex systems we apply time-resolved x-ray techniques allowing us to obtain a real-time picture of the structural dynamics of chemical and biochemical systems in the crystalline and in the liquid phase.

Common for all time-resolved x-ray experiments is the applied pump / probe scheme, where an optical pump-laser initiates a reaction whose structural time evolution is then investigated by x-ray probe pulses at various time delays. The x-ray photon-in / photon-out techniques are based on diffraction or spectroscopic techniques like near edge spectroscopy or x-ray emission spectroscopy. Meanwhile x-ray spectroscopic techniques probe the local environment around specific atoms in a molecule such as orbitals, crystallographic experiments (monochromatic or Laue) reveal the structure of the bulk of periodic systems. Time-resolved diffuse x-ray scattering experiments give information about the structure of liquids.

In the current contribution we will present our latest efforts in that respect. We will reflect capabilities and limitations of state-of-the-art x-ray techniques for the investigation of two different kind of chemical reactions in complex environment: addition reactions in the solid state and dissociation reactions in the liquid state. We will discuss our current status in reaching this goal (proof-of-principle experiments with free electron laser radiation [1], [2]) and how the investigation of chemical reactions benefits from pulsed synchrotron radiation and Free Electron Laser science as they have been performed at the FLASH facility and at LCLS.

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Keywords: FEL research, ultrafast chemistry, structural dynamics

MS.45.4

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Single-pulse laue TR diffraction: Methods, results and use of QM/MM theory

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