Center for Nanoscale Materials, Advanced Photon Source

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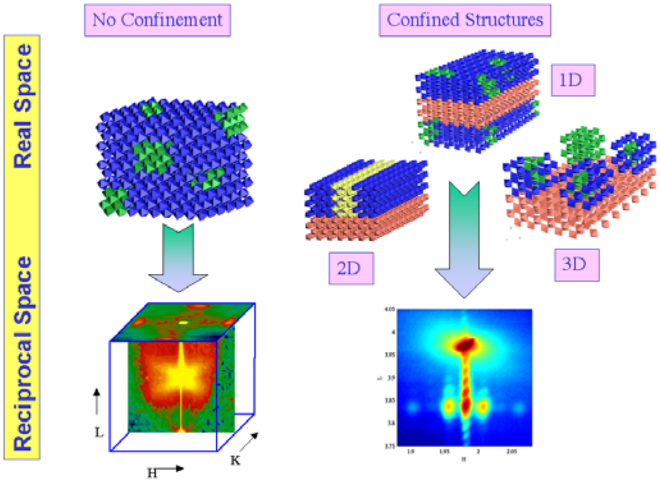
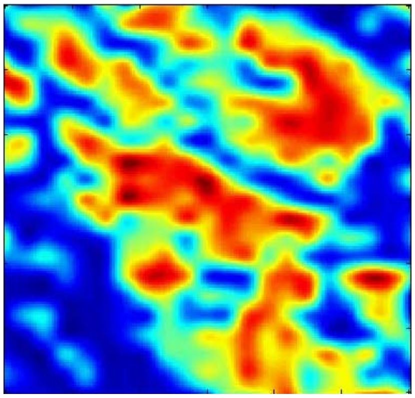
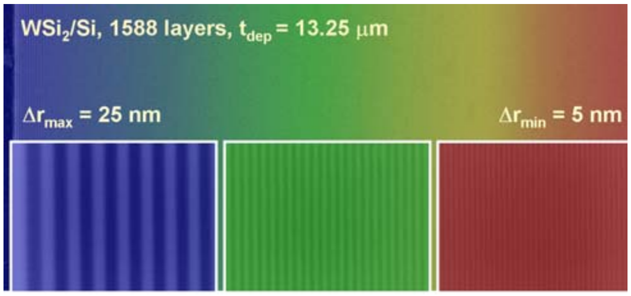
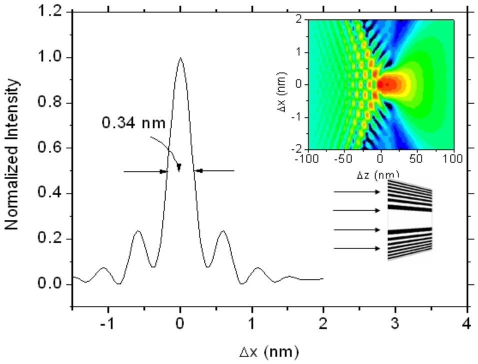
Multilayer Laue Lens Microscope: Nanometer Imaging with Hard X-rays

Requested Funding: $3,855,000









Executive Summary

The possibility of imaging at near-atomic resolution using short-wavelength x-rays has been a dream ever since the nature of x-rays was first understood nearly 100 years ago. With the current availability and future plans for high-brilliance, hard x-ray facilities such as APS, NSLS-II and LCLS, the remaining challenge in achieving this dream has been the availability of x-ray focusing instrumentation with sufficient resolution. Indeed, the development of x-ray optics for 1-nm focusing has been identified as a strategic goal for the next decade by the scientific community.[1] A 1 nm hard x-ray beam will enable fundamental advances in understanding structure and composition at the level of individual atoms, for single nanoparticles, nanocomposites, and functional interfaces in realistic environments such as those required for catalysis; the underlying principles governing self-assembly and synthetic processes for nanomaterials; and dynamics in diverse classes of systems.

CNM and APS have made recent major advances towards this goal and have successfully demonstrated a breakthrough optics concept called the Multilayer Laue Lens (MLL), that has both high resolution and high efficiency[2–7]. Based on this outstanding progress, we propose to develop a microscope that can appropriately manipulate both a sample and our novel optics with the required accuracy and stability for sub-10nm x-ray microscopy. This instrument will push the frontier beyond the current world-leading 30nm-resolution x-ray microscope at APS sector 26 into a regime that is critical for nanoscience, materials science, life sciences and beyond.

Multilayer Laue Lens Microscope

We propose to develop a Multilayer Laue Lens Microscope (MLLM) and operate it at the CNM Nanoprobe Beamline at the Advanced Photon Source (APS). The MLLM will be the first instrument to produce an x-ray focus of below 10 nm, and correspondingly enable a wide range of high-impact scientific applications that are mission critical for the APS and CNM.

Multilayer Laue Lenses (MLL) have been developed at Argonne National Laboratory to focus x-rays to the nanometer scale.[2-3,6,7] Theoretical studies show that these x-ray optics can in principle achieve spot sizes of one nanometer or below.[4] Experimental results with partial MLL structures have to date demonstrated efficient focusing to a line with a width of 16 nm at photon energies of 20 and 30 keV.[5] Full MLL structures aimed at focusing to below 10 nm are under development, and an MLL positioner that allows two-dimensional focusing has been developed.

The MLLM will integrate the MLL optics with an appropriate positioning structure, instrument controls and detectors to yield a fully functional highest-resolution x-ray microscope. In the initial phase, the MLLM will allow focusing of hard x-rays into a focal spot with a size of below 10 nm. Recent advances in the development of MLL optics are expected to allow focusing to a spot size of 5 nm or below in the near future. The MLLM will be based on technology developed for the Hard X-ray Nanoprobe, and integrate an MLL optics nanopositioning stage developed at ANL. The MLLM will use hard x-rays with photon energies between 10 and 20 keV. This energy range is well suited to use x-ray fluorescence, x-ray diffraction and coherent x-ray diffraction imaging for the study of nanoscale objects and structures.

The concept for the MLLM is an extension of the existing CNM Hard X-ray Nanoprobe Instrument, now installed at sector 26 of the Advanced Photon Source. In particular, 8 laser interferometers will be used to encode specimen and optics positions. Active vibration feedback, as demonstrated in the Hard X-ray Nanoprobe Instrument, will be used to maintain the position of optics and specimen. The optics module will consist of a nanopositioning stage that positions two linearly focusing MLL structures into a combined focal spot. This MLL optics nanopositioner allows alignment of each MLL structure to the Bragg angle corresponding to the photon energy used, and alignment of both of them orthogonally to each other. Due to dynamic diffraction, optimized MLL structures have a diffraction efficiency of well above 30%, yielding a total diffraction efficiency of 20% - 35%. Depending on the outermost structure width, such MLL structures can provide a spatial resolution of 5 nm or below.

Acquisition Strategy and Cost

In developing this proposal, the CNM has interacted quite closely with XRADIA Inc., the company that constructed the existing Nanoprobe Instrument based on prototype Argonne designs. The proposal cost estimate is based on a budgetary quotation from XRADIA for the MLLM base system, including supports, scanning system and controls, and Optodyne laser encoding system. We will explore procuring these items from other vendors as well, with the goal of executing a best-value procurement to optimize price/performance. Argonne will provide the MLL optics nanopositioner and MLL optics. The cost of the MLL optics nanopositioner is an engineering estimate based on fabrication costs for the existing ANL MLL optics nanopositioner prototype. MLL optics will be provided by the existing staff effort on this topic in the CNM and APS. Argonne overheads of 10% must be added to quotes for capital procurements; fabrication costs and staff effort estimates are fully loaded. Detectors will be shared with the existing Nanoprobe Instrument.

MLL Microscope Components		**K$**
Equipment		
MLL Microscope base system	Base System Price	$820
	Modification to 8-axes Optodyne laser encoder	$400
	Vacuum System	$250
MLL optics nanopositioner	Prototyping, engineering and fabrication costs	$250
Auxiliary components	Auxiliary detectors, electronics, slits	$150
CCD detector		$60
EDS detector		$235
MLL deposition	Materials	$150
Effort		
MLL positioner - design	½ FTE engineer, 2 years	$300
Optics deposition	Optics scientist, 1 FTE/yr, 3 years	$450
Optics postprocessing	Optics, SA, 1 FTE/yr, 3 years	$240
System Integration	Instrumentation scientist, 1 FTE, 3 years	$450
Total Equipment		**$2.415**
Total Effort		**$1.440**
Total Proposal Request		**$3.855**

Procurement of the MLLM base system is estimated to take one year. Procurement of positioners and controls and assembly/testing is expected to take 6 months, in parallel with procurement of the MLLM. Integration and commissioning of the MLL is expected to take 6 months. MLL optics will be fabricated by the Optics Fabrication and Metrology group of the APS. Partial MLL structures with a resolution limit of 12 nm are available at this date. Development of MLL structures with a resolution limit of below 10 nm is in progress. R&D aimed at achieving MLL structures with a resolution of below 5 nm is ongoing, and continue for a year beyond procurement, integration and testing of the instrumentation hardware.

Scientific and Technical Justification

As the first instrument to focus hard x-rays to the sub-10nm scale, the scientific impact of the MLLM will be very broad. Many of the most intriguing challenges in materials today involve understanding the nanoscale inhomogeneities in crystalline, electronic or chemical structure that arise from competing interactions. In general, these cannot be imaged with the 30nm resolution of the current CNM Nanoprobe Instrument. These challenges include the structure of the cores of magnetic vortices, the electronic or magnetic structure of individual defects, or ground and excited state inhomogeneities near phase transitions. One example of the latter is the emergence of metallic transport in correlated or Mott insulators in which metallic and insulating phases can coexist and even compete for space in the material near the phase transition, with the insulating state gradually winning the competition as temperature is lowered. X-ray methods with high spatial resolution would be an excellent probe to study the dynamics of these charge and structural inhomogeneities with nanometer scale precision. Nanoscale inhomogeneities are believed to be at the heart of this, and of many other important phase transitions such as superconductivity and antiferromagnetism. In another example, calculations indicate that with a 5 nm beam of hard x-rays it will be possible to measure the fluorescence from one atom (~ 100 cps) embedded in a real material such as silicon or to detect its position. Similarly, we expect the landmark sub-10nm x-ray resolution provided by the MLLM to dramatically benefit phase sensitive X-ray imaging methods. Out of proportion to the straightforward gain in instrument resolution one expects with other techniques, when utilizing coherent x-ray diffraction the resolution gain provided by the MLLM has the potential to make possible the study of individual nanocrystals with sensitivity approaching single atoms (Fig. 1). Using coherent diffraction techniques, this will make possible the study of nanocrystal strain and structure induced by the presence of single defects, substitutions, or interfaces, elucidating the fundamental physics behind nanoparticle behavior. This in principle allows fundamentally new areas of physical sciences related to nanocrystal structure and response to be observable on a near-atomic scale via phase-retrieval methods.

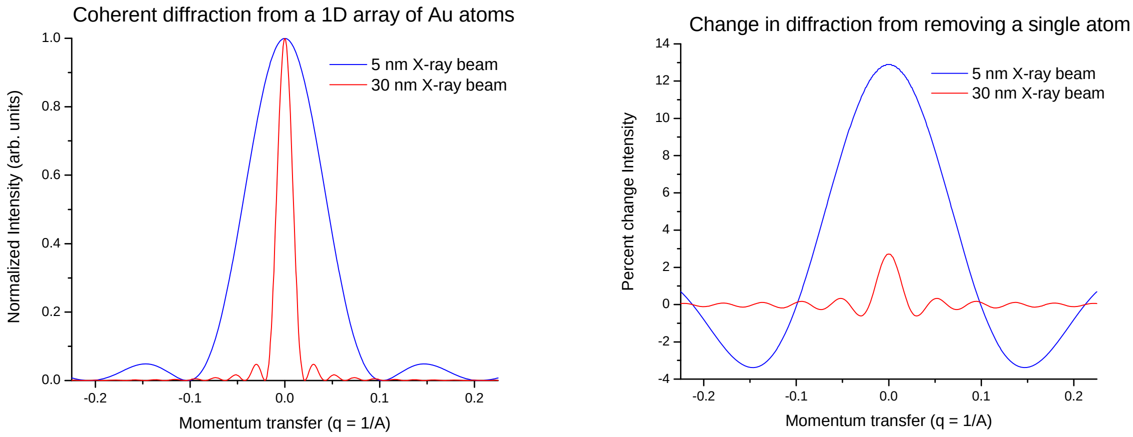


Figure 1 (left): Simulation of the coherent diffraction pattern from a 1D array of atoms with 0.4nm spacing, representing a Au nanowire. (right) Differential response of the coherent diffraction pattern upon removal of a single atom in the wire, showing a six-fold increase in sensitivity when going from a 30nm beam to a 5 nm beam. The increase in contrast is such that a 5nm beam in principle enables observation of single-atom defects in nanoparticles.

With such sensitivity it will also be possible to measure the valence [electronic] state and perhaps even the magnetic state of a single impurity atom in a host semiconductor or metallic nanoparticle. It may even prove possible to measure the Kondo state of a single magnetic impurity.

The increase in resolution coupled with the beamline's existing diamond polarizer will be transformative for studies of nanomagnetic systems, where examination of small buried structures is essential for understanding fundamental behavior. For example, the interior magnetization of layered nanoscale systems that are of order 20 nm thick cannot be studied using traditional magnetic microscopy techniques. For instance, magnetic vortices, in which the magnetization curls around the outside edge of the structure in order to minimize magnetostatic energy, are formed in cylindrical nanoparticles of such dimensions. Current measurements of these systems simply show the existence of a vortex core with the magnetization forced out of the plane. In order to examine the details of core structure, a probe with 10nm or smaller resolution is needed.

Additionally, due to the penetrating nature of hard x-rays, the MLLM will enable the study of buried structures and *in-situ* measurements, including glass-crystalline transformations in confined geometries and charge transfer processes in nanodevices. Magnetic and electric fields can be applied without affecting the x-ray focus, and dynamics at specimen-intrinsic times scales can be studied using the pulsed structure of the x-rays provided by APS. These features will allow study of the intrinsic time response of nanostructured ferroelectrics, the dynamics of grain boundaries, and the behavior of nanoscale complex oxides near phase transitions. Applications will also extend to the nano-bio interface, where the interaction of individual nanocomposites with subcomponents of biological cells can be probed.

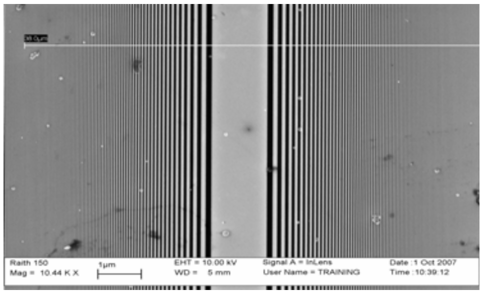
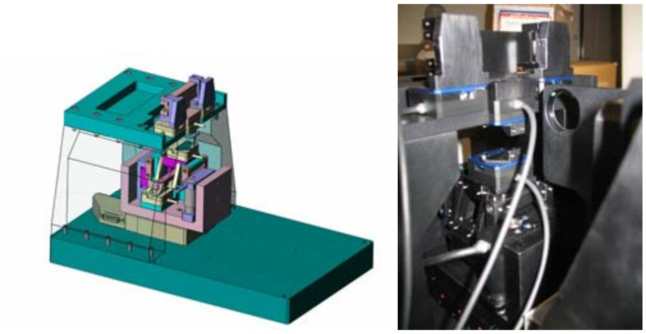


Fig. 2. SEM image of a full linear MLL with an aperture of 40 μm and a smallest structure of 4 nm
(Courtesy Al Macrander, APS)

Significant steps towards nanofocusing have been made by the proposers through an internally funded (LDRD) project at ANL. The efforts have included (i) a theory thrust, (ii) a thrust on optics fabrication and (iii) a thrust on mechanical positioning. The theory thrust has led to the development of a new theoretical approach showing that MLL's will efficiently focus hard x-rays to a spot of 1 nm or smaller.[6] The optics thrust has so far yielded partial linear MLL structures with an outermost structure width of 5 nm and a resolution limit of 12 nm. A line width of 16 nm has been measured using these structures. Development of depositing full linear MLL structures with a resolution limit below 10 nm has shown promising early results (Fig. 2). Initial experiments with the deposition of wedged MLL structures, which are designed to locally satisfy the Bragg condition across their full aperture and which are required to achieve x-ray focusing to below 5 nm, have also been successful[7]. The development of advanced positioning systems has yielded a first device for positioning two MLL structures orthogonally to each other and in close proximity (Fig. 3).



A) B)

Fig. 3. Prototype MLL positioning device.
(A) Engineering view of 8-axis positioning system for two orthogonally aligned MLL structures. (B) Photo of the prototype MLL positioning device.

Usage of the Instrument

The MLLM will be operated as a user instrument at the Nanoprobe Beamline, Sector 26 at the Advanced Photon Source. The configuration of the Nanoprobe endstation allows use of either the Nanoprobe instrument or the MLLM. The MLLM will provide two major modes of data acquisition, x-ray fluorescence mapping and x-ray diffraction. The latter will include in particular coherent diffraction imaging. Access to the instrument will be provided through the CNM proposal system. Time allocation between the Nanoprobe instrument and the MLLM is not fixed, and will take into account user request for each individual system. We assume that a significant fraction of users studying nanoscale system will be interested in highest spatial resolution, and estimate a usage of 1000 – 2000 hour/year of beamtime, corresponding to 15 - 30 users.

Appendices: References, Curricula Vitae, Quotes

References

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[6] Members of the ANL team received a 2005 R&D 100 Award for developing the multilayer films for MLL structures.

[7] R. Conley, C. Liu, J. Qian, C. M. Kewish, A. T. Macrander, H. Yan, H. C. Kang, J. Maser, and G. B. Stephenson, "Wedged Multilayer Laue Lens," to appear in *Review of Scientific Instruments* (2008).