# Are Atom-sized X-ray Experiments Possible?

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**Abstract.** The success of advanced microbeam facilities at third generation synchrotron sources have inspired us to ask ultimate questions such as how small an x-ray beam diameter can be made. With the hope of more brilliant Energy Recovery Linac or X-ray Free Electron Laser sources due to arrive in the next decade, it appears possible to think of fluorescent x-ray experiments that can be performed on even a single impurity atom in a silicon wafer, for instance. Not all x-ray optical developers are yet convinced, however, so there is critical need to assess whether in principle this can really be done or not. We are optimistic that 1 nm diameter x-ray beams can be made of sufficient flux from future sources or even demonstration experiments at lower count rates from 3rd generation sources if it turns out to be worthwhile to actively develop optics and methods that vastly exceed the current x-ray microbeam capabilities.

## INTRODUCTION TO THE IDEA OF SINGLE ATOM X-RAY EXPERIMENTS

A recent Applied Physics talk at Cornell University by Paul Voyles of Bell Laboratories discussed imaging single Sb atoms in a 20 Angstrom thin silicon wafer using Transmission Electron Microscopy (TEM) with 200 keV electrons[1]. At the high dopant concentrations needed for the smallest transistors, these atoms can form electrically inactive clusters and not function properly. As a result of hearing this talk, we began to ask ourselves: could we do this same measurement just as well with x-rays and also use thicker samples? Could we focus an x-ray beam on a single atom and extract x-ray data with all the power of techniques that have been built up in the x-ray community over the last 100 years or so? The purpose of writing this paper is to stimulate the microbeam community to prepare for new opportunities ahead and to help start the dialog about whether it will be feasible to conduct x-ray experiments on a single atom within the framework of the next 10 years of development.

Below we review important fundamentals and conclude that single atom experimentation with x-rays will require careful development and lots of work and investment. At this point in time, there is also not a consensus in the developer's community as to ultimate feasibility, so even conclusions that seem promising today will need to be critically examined and tested over the coming years.

To begin to explore the prospect of conducting an x-ray experiment on a single atom requires we look into the areas of experimental design, x-ray sources and then x-ray optics. The critical questions are: 1) can the beam be focused small enough? 2) will there be enough x-ray intensity? and 3) can radiation damage be overcome? Below we lay out some general ideas and then draw the conclusion at the end that single atom experiments may be possible, but our tentative conclusion will need further study and investigation to fully clarify limits that to date have not been well explored.

First, we set the stage by discussing the concepts of why we wish to conduct such small beam experiments followed by assessments of what we will have to develop to make optics and x-ray sources suitable for these experiments.

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## FIRST SINGLE-ATOM EXPERIMENT IDEAS

Microbeam experiments at 3rd generation synchrotron sources are a big success[2], especially for experiments where the beam size is made to be of order 1 micron or less. The drivers for this success are the small synchrotron source size, its high brilliance, and the continuing development of microbeam x-ray optics. Out of this fine success story comes the concept of fundamental limits to the size of an x-ray beam. Are there any limits? At this point, this is question is mainly of academic interest, but in today's climate of "nanotechnology", there is lots of interest to look at smaller and smaller objects with x-rays, all the way down to the limits of the size of molecules and even one single atom! What has held us up from presently reaching this limit is that no x-ray optics have been made to focus a beam to atom size nor is there presently enough intensity to make practical experiments in a reasonable amount of time. We will discuss these two points further below. But why interest in focusing an x-ray beam to atom size? After all, by powder or single crystal diffraction methods, we can already take larger beam sizes and extract atomic resolution detail based on multiple copies of a unit cell with sub-Angstrom resolution!

In the case of the Voyles experiment introduced above, if we had the x-ray sources and optics discussed below, we might well be able to directly form x-ray fluorescent images to locate the atoms with two dimensional scans or even in three dimensions if we use tomographic methods. A direct image of a two-atom cluster might be possible as well. With enough intensity, we could also do spectroscopy on each atom, most likely learning enough about the local chemical environment from near edge studies so that conclusions could be drawn about its electrical activity in a semiconductor. This might be useful for when modeling the properties of semiconductors in the limit of vanishingly small size. The novel part is that the data would be taken from a single atom and not averaged over many similar atoms of a larger structure, as is the case for all of the x-ray spectroscopy measurements taken to date. Other techniques could be very viable also, such as x-ray fluorescence holography where the local environment could, for the first time, be probed around just one (or several) atom(s)[3].

In the larger context of ultra-small beams and very thin samples, however, Electron Microscopy is generally the method of choice for really focusing electron beams down to the size of about one Angstrom[4]. In many ways, electron probes are preferred as they are more well developed, instruments are readily available, and the cross section for electrons interacting with an atom are relatively high, compared with x-rays. On the other hand, we can expect to find complementary situations where the properties of x-rays are really needed such as when the sample is too thick to be accurately imaged with electrons.

## X-RAY SOURCE QUALITIES

To make useful x-ray beams for a nanometer dimension probe from a partially coherent light sources such as a storage ring, we need sources of very high brilliance and small source size that can be further demagnified. For instance, a spot size of 30 nm is planned for the Hard X-ray Nanoprobe beamline at APS when it is commissioned a few years from now[5]. Other microfocus beamlines from 3rd generation synchrotrons should be able to move into this territory as well. This is about the current conceptual limit for the next several years. With top-up modes of operation and few nm-rad horizontal emittances, these storage ring machines are poised to push the frontier to substantially smaller beamsizes than we work with today.

To do even better, we will need even more brilliant x-ray sources that are linac based. The proposed Energy Recovery Linac (ERL)[6] can make a round source of 3.7 microns in size (rms) that can be demagnified (in-principle) down to a nanometer beamsize (see optics discussion below). The ERL machine will be an ideal microbeam source not only because of it's factor of 10 to 100 more brilliance than storage rings, but also because its small source size will be round, a feature that helps to make a simple round spot size behind nearly perfect optics that could conceivably deliver as many as $10^{10}$ to $10^{12}$ x-rays/sec/nm$^2$, enough intensity to conduct experiments with a very reasonable count rate of up to $10^4$ to $10^6$/sec from a one impurity atom[7]. The TESLA[8] and LCLS[9] XFELs will also have an average brilliance that significantly exceeds that of storage rings and additionally produce fully coherent beams. Thus optics such as zone plates will have a beamsize determined by the dimension of the outer zone and not just by the source size. One drawback for some microbeam XFEL experiments, however, may be the low duty cycle of the machines. Will the highly focused peak brilliance with a low duty cycle so heavily damage the sample that it alters its structure? For the ERL case, our group has concluded that for the nearly 1.3 GHz pulse rate, we are no worse off than the TEM experiment of Volyes and are hopeful that samples based on silicon will adequately survive such a focused CW beam without significant damage[10].

## X-RAY OPTICS NEEDED

Next we access whether 1 to 10 nm beam sizes can in principle be achieved using zone plates, nanofabricated tapered waveguides, etc. Obviously these kinds of actual x-ray optics that are currently beyond the state-of-the-art, but we might deem them possible in the near future after much more development work. Thus we push to think about design limits just that don't violate the laws of physics even though we can't currently reach this level of fabrication performance today. Two optical choices stand out: waveguide or tapered capillaries to reach 10 nm size[11] or zone plates[12] to make a 1-3 nm beam size[13],[14] at 5 to 10 keV. The authors of Ref 11 suggest that a 10 nm limit "appears to apply to all x-ray focusing devices of similar efficiency", a conclusion that needs review, further consideration and testing by the x-ray optics community.

## CONCLUSIONS

Single atom x-ray experiments appear conceivable in the next 10 year period, but much work remains to be done by the x-ray microbeam developers to elucidate the fundamental limits of this technology and to help find ways to fabricate optics and conduct experiments on such a fine nm scale. The prospects of working with these extremely brilliant nanobeams of x-rays will be an important theme of future high-brilliance x-ray sources.

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## REFERENCES

[1] Voyles, P. et al., Nature 416, 826-828 (2002).
[2] Riekel, C. Rep. Prog. Phys. 63, 233-262 (2000).
[3] Tegze, M. and Faigel, G., Nature 380 49-51(1996).
[4] De Graef, M., Introduction to Conventional Transmission Electron Microscopy, Cambridge University Press, Cambridge, 2003, pp. 1-740.
[5] Brian Stephenson, personal communication.
[6] Gruner, S.M., Bilderback, D.H., Nucl. Instrum. & Methods A 500, 25-32 (2003).
[7] Banerjee, S, Bilderback, D.H. and Huang, R, "Feasibility of Single-Atom X-ray Fluorescence Imaging from an Energy Recovery Linac Source of Synchrotron Radiation", unpublished paper from Applied & Engineering Physics 490 class (May 2002) posted at http://erl.chess.cornell.edu/papers/Banerjee2002.pdf
[8] TESLA-Technical Design Report, DESY 2001-011, ECFA 2001-209, CDROM, March 2001 See

http://tesla.desy.de/newpages/tdr_update/start.html

[9] Linac Coherent Light Source (LCLS) Design Study Report, SLAC-R-521, Dec. 1998. The report can be found at http://www-ssrl.slac.stanford.edu/lcls

[10] Huang, R and Bilderback, D.H., unpublished calculation.

[11] Bergemann, C., Keymeulen, H., van der Veen, J.F., "Focusing X-ray Beams to Nanometer Dimensions", manuscript submitted to Phys. Rev. Letters (2003).

[12] Kurokhtin, A.N., Popov, A.V., J. Opt. Soc. Am. A 19, 315-324 (2002).

[13] Brian Stephenson, personal communication.

[14] Yun, W., personal communication. More background information can be found at http://www.xradia.com