Facility Highlight

Impact of a Future Energy Recovery Linac X-ray Source on Nanoscale Science

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The performance characteristics of modern 3rd generation synchrotron X-ray sources are approaching fundamental physical limits set by the equilibrium dynamics of particle storage rings. But this limit can be exceeded using Energy Recovery Linac technology with exciting consequences for future nanoscale X-ray science [1]. A "white paper" last updated on November 30, 2000 describes the ERL concept [2] and includes brilliance and flux comparisons to existing and some proposed future light sources. The spectral curves have since been updated for higher performance [3] as we have learned more about the ERL design process.

First ever: small, round synchrotron X-ray source. Detailed machine designs [4] developed by Cornell University's ERL prototype project staff demonstrate that an ERL would be an essentially diffraction-limited X-ray source for photon energies less than or equal to 12.6 keV and with a (circular) emittance of 0.008 nm-radians. These parameters are outstanding and offer the opportunity to demagnify a 2 micron (rms) round source to 1-10 nm circular beam waists for focused X-ray beams, but with intensities comparable to current 3rd generation synchrotron beam lines. Such an ERL type of X-ray source would enable the application of essentially all existing X-ray characterization techniques to individual nanoparticles (e.g., 20-50 nm) and perhaps even atoms. For example, fluorescence detection and spectroscopy of individual impurity atoms in a clean silicon wafer might become possible. A variety of ultra-fast (sub-picosecond) and coherent X-ray imaging techniques also become possible on very small specimens. We explore in this article a few of the interesting future nanoscience areas that may be feasible with an ERL X-ray source.

To put future X-ray microbeam developments in perspective, let us consider some examples at different length scales and reflect upon the science possible with different X-ray probe sizes. Fig 1 shows such a range of probe sizes from the centimeter scale down through the atomic scale. The middle example, involving diffraction from individual micron-sized

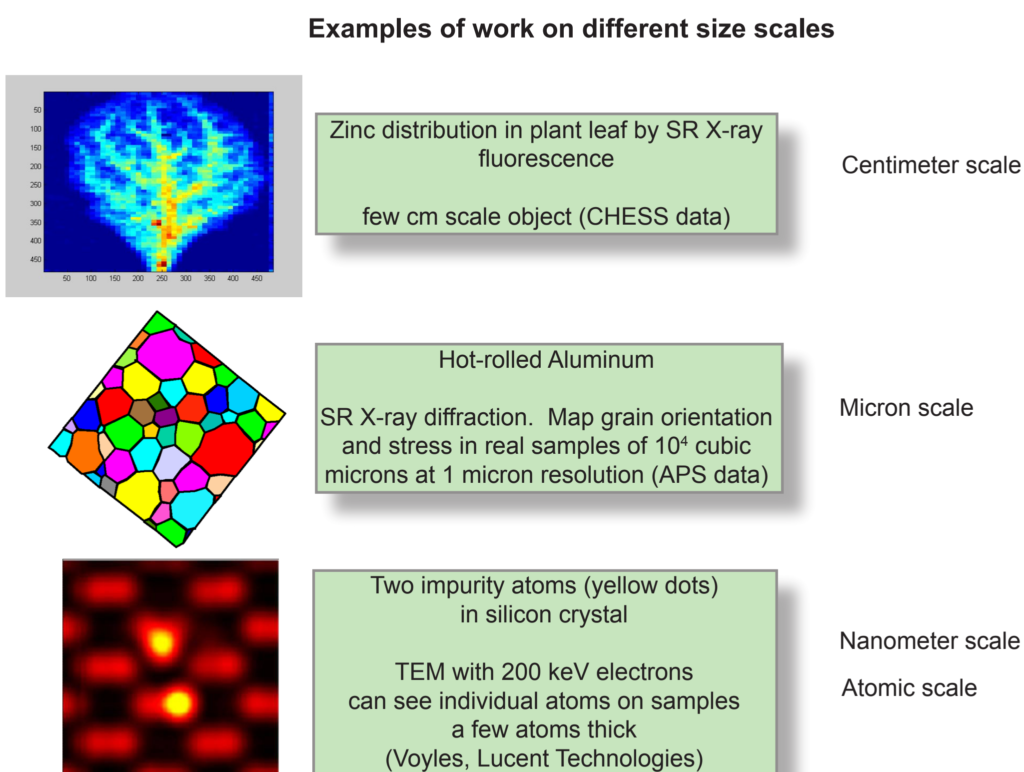

Fig 1: Examples of microbeam work on different length scales. A plant leaf image in Zinc Kα X-rays shows where a heavy metal has accumulated in the "arteries" of a leaf a few cm in size (image from CHESS). Going smaller in size, an X-ray grain orientation map has been made on micron-sized aluminum domains at the Advanced Photon Source. This experiment really requires a third-generation type X-ray source such as the APS as it is an example of a really brightness-limited situation. Each of the colors shows a grain of different orientation and where the grain borders are located. The last image is made from 200 keV electrons (not X-rays) from a TEM microscope [5] as imaged through a 20 Ångstrom thick doped silicon wafer. The image shows individual silicon atoms in a regular lattice with two antimony atoms in the middle (yellow color). Making X-ray beams of the order of magnitude of atomic dimensions would be very useful for a wide variety of experiments.

grains in a metal, represents the present state-of-the art. The much smaller beams possible with an ERL would allow this experiment to be done with sub-100 nm-sized grains, or, just as importantly, to determine the strain gradients across 100 nm-sized grains. The third example in Fig 1 illustrates single-atom imaging. Presently, atomic-scale imaging is done with electron microscopes because electron beams can be readily focused to nanometer dimensions. However, electrons have difficulty penetrating thick samples without multiple scattering effects that degrade the image, so electron microscope samples are rarely thicker than a micron. A nanometer-sized ERL X-ray beam would allow atomic imaging of atoms buried in thick samples.

Future nanoscience beamline. Fig 2 shows a schematic of an instrument for making and using nm diameter X-ray beams. If appropriately efficient optics could be devised, up to 10^{12} X-rays/sec/nm^2 would be present in the focus of this instrument [6]. With such a large flux in such a small spot, X-ray fluorescence from a single atom would yield as much as 10^6 X-rays/sec, a number suitable for single atom imaging or EXAFS experiments.

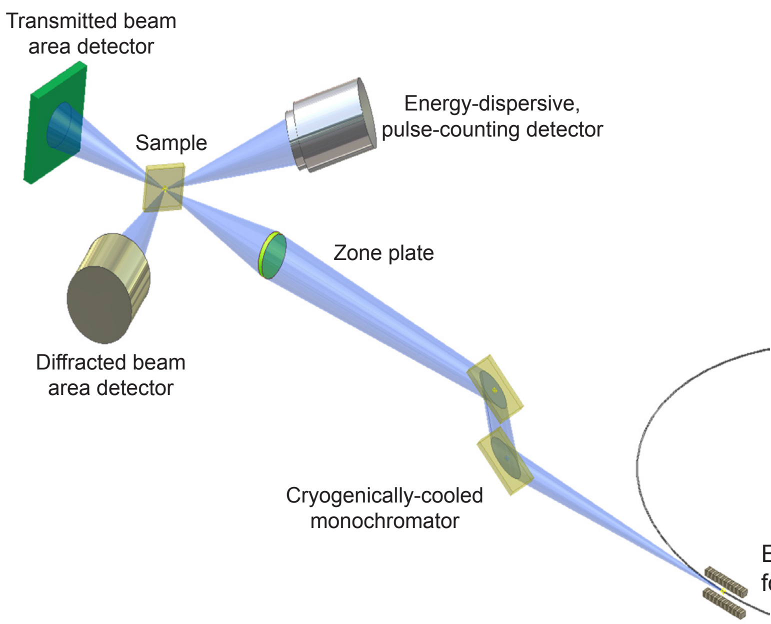

Fig 2: Schematic diagram of a nano-probe beamline on an ERL machine. The combination of a 1 m long undulator with a 2 micron (rms) round source and a demagnification of 2000 would make a 1 nm beam diameter possible, assuming the development of the requisite optics.

These types of experiments might be on the threshold of possibility with 3rd generation storage ring sources, but at greatly reduced flux. For example, the2-ID-D station at the APS for nanodiffraction achieves a focus of 150 nm x 150 nm from a zone plate behind a silicon (111) monochromator and produces a photon flux of more than 10^9 X-rays/second/0.01% bandwidth over a 5.5 to 30 keV energy range [7] with a flux density gain that exceeds 50,000. At SPring8, a similarly equipped beamline, 20XU, made a 120 nm x 130 nm beam with a flux of 2.6 x 10^7 X-rays/sec at 10 keV in first-order diffraction [8]. Going to third-order with 8 keV X-rays produced a 50 nm beam size with a measured flux of 1.7 x 10^5 X-rays/sec. There are more recent reports of achieving even smaller beams with KB mirrors [9] of 36 nm x 48 nm at 15 keV (with estimated flux [10] of 5x10^8 X-rays/sec), and zone plates [11] of 58 nm x 58 nm at 8 keV (with estimated flux [12] of 1x10^8 X-rays/sec).

At the ESRF, a 2-dimensional waveguide [13] has made a 13 keV coherent X-ray beam of 69 nm x 33 nm with a measured flux of 2x10^5 X-rays/sec and a gain of 70. Refractive lenses look promising from a theory point of view [14] to make beam sizes down to 2 nm. A recent test at the ESRF produced a 50 nm x 50 nm beam at 21 keV with 1.6 x 10^8 photons/sec using silicon refractive lenses in a crossed 90 degree geometry behind a silicon (111) monochromator [15].

Transmission multilayered films [16] have recently produced a 29 nm 1-dimensional line focus at APS at 19.5 keV with 45% efficiency [17]. The hope is to eventually take two of these optics together to form a 15 nm x 15 nm beamsize with high efficiency. Overall, the quality of the X-ray optics is improving considerably, so smaller beams at higher efficiencies can be expected within the next several years. However, the full X-ray power of an ERL will be needed to make such nanobeam experiments truly practical on a few nm x few nm scale along with X-ray optics expressly designed for this purpose. An instrumental layout of a nanoprobe, Fig 2, shows a zone plate as the focal element for making a small X-ray beam, but mirrors or refractive X-ray lenses may prove to be more suitable. A variety of detectors should be available for collecting the diffraction and spectroscopy information from a sample.

A recent review by Larson and Lengeler [18] highlighted "the rapid progress that is ongoing in the development of hard X-ray microscopies with three-dimensional spatial resolutions ranging from micrometers to nanometers. The individual articles (that follow in the MRS Bulletin) provide a crosscut of developments in hard X-ray projection tomography microscopy for imaging density and chemical fluctuation in crystalline and noncrystalline materials; large-angle diffraction-based, spatially resolved imaging of local structure, orientation, and strain distributions in crystalline materials; and emerging coherent diffraction imaging for nanometer-range Fourier transform imaging of crystalline and noncrystalline materials."

Fig 3 shows the kind of information that an ERL X-ray nanoprobe might produce on a nm-sized cluster of atoms or individual molecules. Such a probe could produce quantitative atomic-scale structure, strain, and orientation imaging of small crystalline clusters. Using spectroscopy tools, one could increase the fluorescent trace element sensitivity from the present 10^{-19} g to that of a single atom (10^{-24} g) and be sensitive to chemical state via XAFS at ultra-low concentrations.

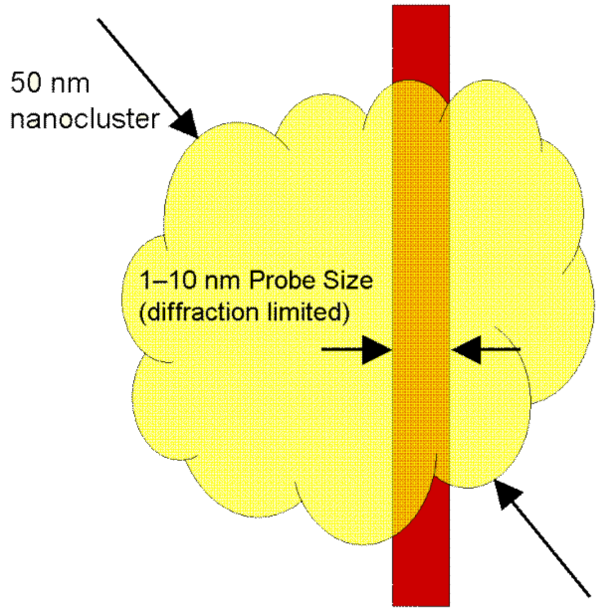

Fig 3: Cartoon of a 1-10 nm diameter beam incident upon a 50 nm sized cluster of atoms (e.g. nanometer size quantum dots [19]). With such a small beam, the internal structure of a cluster could be determined by diffraction, spectroscopy, imaging methods, etc. Since X-rays have the ability to penetrate thick layers, nasty gas environments, etc. (as opposed to electron microscopy), they could find new applications in the world of the ultra-small sample buried in thick and fluid environments.

ERL as a CESR upgrade. The present concept for an ERL on the Cornell campus is to upgrade the present CESR machine by the addition of a long linear tunnel that houses the ERL equipment and generates the ultra-bright electron beam. Fig 4 shows the layout of the machine that is currently under consideration.

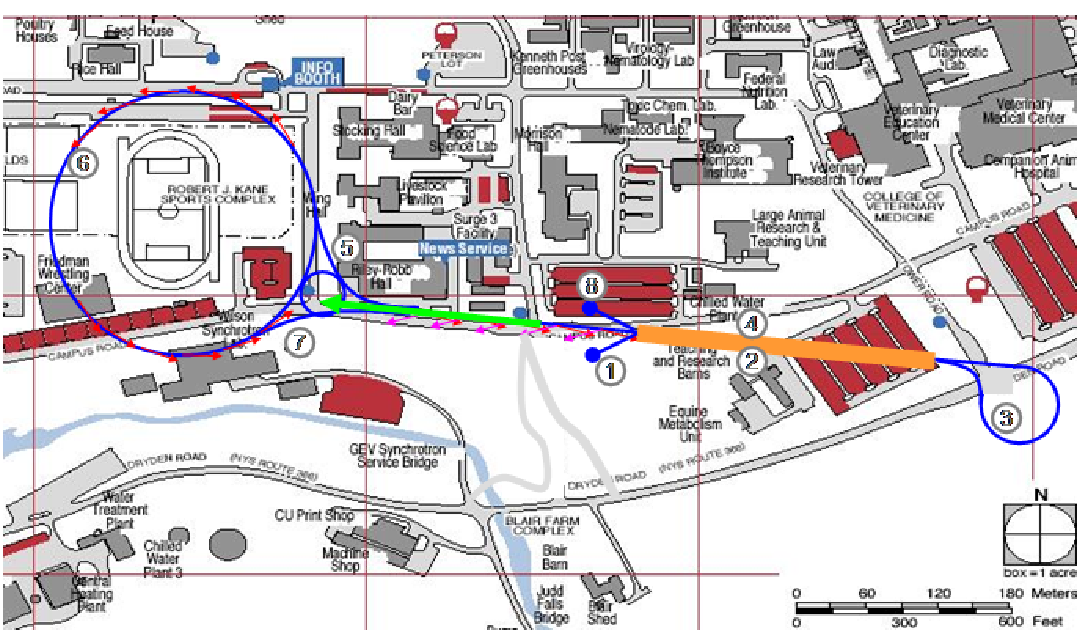

Fig 4: Schematic of one of several ERL upgrades to CHESS under study [20]. The injector (1) feeds the linac that accelerates the beam to half its final energy (2). The beam then loops back (3), is accelerated by a second, parallel linac to the final energy (4), is routed into CESR (5) and (6), is extracted (7), is half energy recovered in linac 2 (4), loops back (3), is fully energy recovered through the linac 1 (2), and finally dumped (8). The green area may be used for future long undulators. The small circle between (5) and (7) allows operation of the ERL independently from CESR for testing purposes.

The spectral brilliance of the ERL in its high coherence mode is shown in Fig 5. A highly brilliant curve means that the ERL will be able to deliver a very high flux of X-rays within a few nm^2 area.

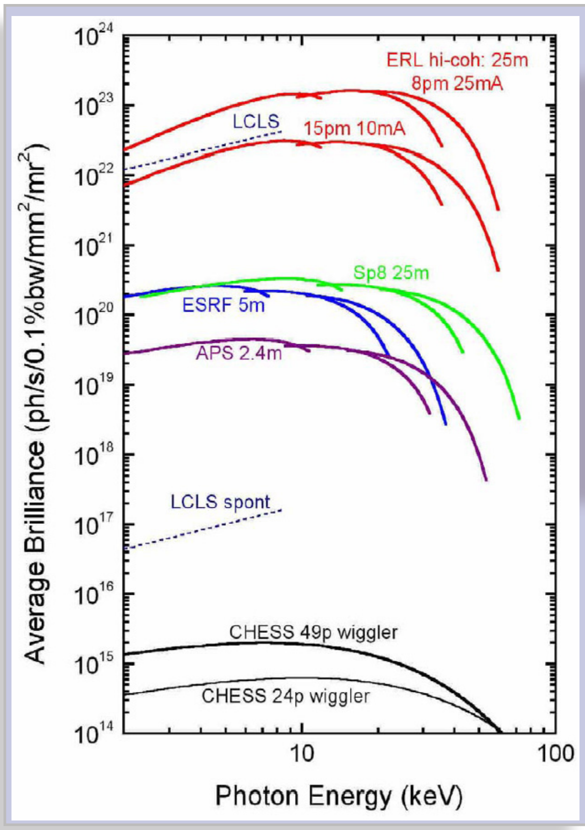

Fig 5: Average spectral brightness of the ERL compared to ESRF, APS, and LCLS [21]. The ERL photoinjector has several operating modes: Initial design goals are flux (0.1nm emittance, 100mA current), brilliance (0.015nm, 10 mA), peak brilliance (0.1 nm, 1 mA, 100fs bunches). Long-term design goals are flux (0.1nm, 200mA), brilliance (8 pm, 25 mA), peak brilliance (0.1 nm, 10 mA, 20fs). Lengths refer to undulator lengths. Note: The LCLS peak brilliance is much higher than any other source, but at a lower repitition rate.

Facility Highlight

Table 1 shows a comparison of existing APS to future ERL parameters. The improvements are sufficiently large that the ERL would be truly transformational!

The 3rd generation machines have been very successful in making smaller beams than initially proposed. We likewise hope that the ERL would make even smaller beams feasible. The beam cross section will be about a thousand fold smaller in area, the coherent flux will be up to 3,000 times more (on longer undulators with slightly larger beamsize). And there will be an opportunity to make experiments using short pulses at up to a 1.3 GHz repetition rate but probably not with the smallest possible beam sizes.

Parameter	APS 3rd Generation Storage Ring	Energy Recovery Linac	Gain Factor
Electron source size in microns rms	239(h) × 15(v)	2(h) × 2(v)	1/900 in area
Micro X-ray beamsize	70 nm to 1 micron	1 nm	70 to 1000
Coherent flux X-ray/sec/0.1% bw	3×10^{11}	9×10^{15}	3000
Pulse duration (rms)	32 ps	<100 fs	more than 320 times shorter

Table 1: Comparison of important microbeam ERL parameters (source size, coherence, and pulse length) with APS.

Future challenges: Radiation damage will certainly become one of the biggest limitations in the use of small beams, especially for soft-matter samples. However, community experience so far has shown that brighter sources catalyze new ideas on ways to mitigate radiation damage (e.g., freeze-drying of specimens). We fully expect this process to continue with an ERL source. Other limitations arise from existing X-ray optics, but there are several X-ray optical groups who are aiming to break the 10 nm hard X-ray barrier by developing better optical components over the course of the next several years.

Conclusions: The ERL source parameters - size, angular divergence, pulse duration and rate - are dramatically better than present 3rd generation storage rings. These parameters should make it feasible to extend to smaller scale all of the existing X-ray techniques that have become standard tools at synchrotron sources such as diffraction, scattering, spectroscopy (including XAFS, XANES, etc.), holography, tomography, imaging [phase contrast, coherent, and fluorescent (with single atom sensitivity, ~10^{-24} gram)]. This capability naturally leads to new scientific opportunities in many fields such as condensed matter, mesoscopic science, biological and chemical dynamics, high-pressure science, etc. The use of ERL microbeams, their coherent beams, and ultra-fast timing structure will lead to new unique experiments that can be expected to transform the way future X-ray nanoscience experiments are conducted.

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