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Coherent X-ray imaging and microscopy opportunities with a diffraction-limited Energy Recovery Linac (ERL) synchrotron source

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Abstract. A proposed Energy Recovery Linac (ERL) x-ray source at Cornell would be a 5-7 GeV synchrotron facility based on the energy-recovery principle with a superconducting linac. Because of its ultra-small round electron source and ultra-short but flexible bunch structure, the ERL has the potential to produce sub-picosecond ultra-bright diffraction-limited hard x-ray beams that are superior to those from existing storage rings, and would enable new scientific experiments that are difficult or impossible to perform today. In this paper, we discuss the transverse coherence properties of the ERL source and show how these properties could benefit various x-ray microscopy applications that require a highly coherent and intense x-ray beam. These applications include full coherent illumination of wide-aperture zone-plate optics for scanning x-ray microscopy, diffraction-limited coherent phase-contrast microscopy and tomography, fully coherent hard x-ray diffraction microscopy, and large-coherence-area x-ray holography techniques.

1. INTRODUCTION

X-ray microscopy has made substantial advances in recent years [1], especially in the hard x-ray regime. These advances can be mainly attributed to two factors: improvements in focusing x-ray optical components and the availability of bright third generation synchrotron sources. Focusing optics have produced sub-100nm focal spot sizes for hard x-rays [2], and condensing optics have reached 50nm spatial resolution [3]. Present-day storage-ring synchrotron sources are essentially diffraction-limited for 8 keV photons in the vertical direction, with source sizes in the 10-20 micrometer range [4]. The combination of these two factors appears to be the key for x-ray microscopic applications using hard x-rays, as can be seen from these proceedings. Compared to other forms of microscopy, x-ray microscopes have the advantage of less radiation damage and greater penetration depth than electron probes, and of much better spatial resolution than optical microscopes [5].

Further advances in spatial resolution in x-ray microscopy require substantial improvements in both x-ray optics and x-ray sources. Although some present-day third-generation synchrotron sources may be diffraction-limited vertically, the horizontal emittance is 10^2-10^3 times larger, resulting in an essentially incoherent hard x-ray source. The extremely anisotropic, flat-beam characteristic also limits efficient throughput for x-ray optics and is thus undesirable for two-dimensional microscopic and imaging applications. Although not every form of microscopy requires high degree of coherence, almost all focusing optics benefit from a diffraction-limited source [6], which is the case for all scanning x-ray microscopes (SXM).

In principle, x-ray microscopy should be able to reach sub-nanometer spatial resolution comparable to the x-ray wavelength. In practice, however, this ultimate resolution may be difficult to achieve by focusing optics only, as current fabrication techniques may reach a limit of 10 nm. To go beyond the 10 nm fabrication barrier, coherence-based x-ray imaging and microscopic methods [7-11] become extremely attractive since the spatial resolution in these methods depend only on diffraction resolution in a coherent diffraction pattern. Such horizon methods require not only a fully diffraction-limited x-ray source for high diffraction resolution applications, but also orders-of-magnitude increase in coherent intensity compared to what is possible at present third-generation hard x-ray sources.

2. PROPOSED ERL X-RAY SOURCE

The recently proposed energy recovery linac (ERL) synchrotron source [12-17] would be an ideal source for ultimate resolution and coherent x-ray microscopic applications. Such a facility proposed at Cornell would be a 5-7 GeV synchrotron source (Table 1) based on closed-loop energy recovery with superconducting linear accelerators and small-gap short-period undulators. It offers significant advantages over storage ring sources, both in terms of the possible x-ray beam quality and, once the technology is developed, cost-effectiveness. The basic idea behind an ERL was suggested long ago for beam colliding machines [18] and the feasibility of operating an ERL has recently been demonstrated with a highly successful free electron laser at the Thomas Jefferson National Accelerator Facility.

Table 1: Preliminary design parameters for the Cornell ERL source.

	ERL High-flux	ERL High-coherence	Compared to ESRF [4]
Beam energy E_G (GeV)	5.3	5.3	6
Beam current I (mA)	100	10	200
Hori. emittance ε_x (nm-rad)	0.15	0.015	4
Vert. emittance ε_y (nm-rad)	0.15	0.015	0.01
$\varepsilon_x\varepsilon_y$ (nm-rad)^2	0.02	0.0002	0.04
Bunch length fwhm τ (ps)	0.3-5	0.3-5	35
Undulator length L (m)	25	25	5
Undulator period λ_u (cm)	1.7	1.7	3.5
Energy @ 1st harmonic (keV)	8.0	8.0	8.0
Average flux F_n (phs/s/0.1%)	$1.5\cdot10^{16}$	$1.5\cdot10^{15}$	$1.3\cdot10^{15}$
Average brilliance B (phs/s/0.1%/mm^2/mr^2)	$1.3\cdot10^{22}$	$5.2\cdot10^{22}$	$3.1\cdot10^{20}$
Hori. divergence fwhm (μrad)	9.1	6.2	26.8
Vert. divergence fwhm (μrad)	9.1	6.2	10.4
Hori. source size s_x fwhm (μm)	103	24.5	879
Vert. source size s_y fwhm (μm)	103	24.5	13.9
Coherent flux F_c (phs/s/0.1%)	$8.1\cdot10^{13}$	$3.1\cdot10^{14}$	$1.8\cdot10^{12}$
Coherent intensity $4F_c/(\pi s_x s_y)$ (phs/s/mm^2/0.1%/1:1 focus)	$1.0\cdot10^{16}$	$6.6\cdot10^{17}$	$1.9\cdot10^{14}$

The basic machine parameters for the proposed Cornell ERL are listed in Table 1, which is taken from Ref. [14]. The product of the vertical and horizontal emittances is important, since the brilliance, and, hence the coherence, scales with the inverse of this product. As can be seen from the Table, the ERL would produce round synchrotron x-ray beams with average brilliance more than one order of magnitude higher than the existing storage rings, making the ERL comparable in this regard to proposed prototype 4th generation x-ray free-electron laser (XFEL) sources. For x-ray microscopy applications, the high coherence ERL would improve the source properties in two ways: by reducing the source size

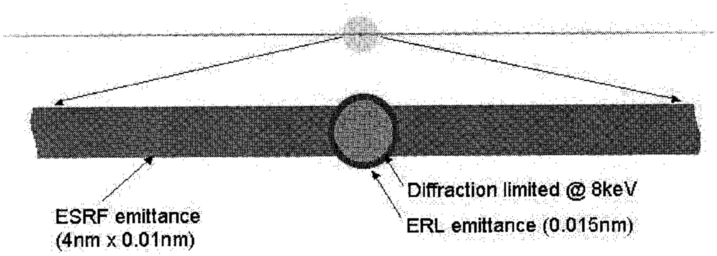

Figure 1. Schematic illustration of transverse emittances of the proposed ERL synchrotron source, compared to the emittances at a third-generation storage ring, ESRF. Also shown is the diffraction limited emittance area at 8 keV for the ERL.

by one-to-two orders of magnitude in the horizontal direction to make essentially a round beam as illustrated in Fig. 1, and by providing more than two orders-of-magnitude higher coherent flux as compared to third generation storage ring sources. The combination of the two factors is reflected in the coherent intensity or flux density at the focal spot assuming a 1:1 focusing ratio, leading to a potentially 10^3 increase in coherent flux delivered onto a specimen.

3. X-RAY MICROSCOPY APPLICATIONS

The intense, ultra-small, round x-ray beam produced by the proposed ERL source would offer substantial advantages for x-ray microscopy applications. Here we briefly discuss several of these potential advantages.

(1) *Full coherent illumination of wide-aperture optics*:

A typical scanning x-ray microscope uses a Fresnel zone plate to demagnify an x-ray source. The effect of coherence illumination of a wide-aperture zone plate has been studied extensively by Winn *et al.* [6]. It has been found in order to achieve the best possible spatial resolution, the usable wavelength normalized source phase space area $2\pi\varepsilon/\lambda$ has to be limited to <1, where ε is the source emittance. The corresponding source, with $\varepsilon = \lambda/2\pi$, is usually referred as an *almost diffraction limited* source, as compared to $\varepsilon = \lambda/4\pi$ for a fully diffraction limited source. Because all existing hard x-ray synchrotron sources are far from the diffraction limit, it is often necessary to insert an aperture in front of the zone plate optic to select a small horizontal portion of the undulator radiation in order to achieve the desired spatial resolution for a SXM. This trade-off of course limits the available x-ray intensity to a sample.

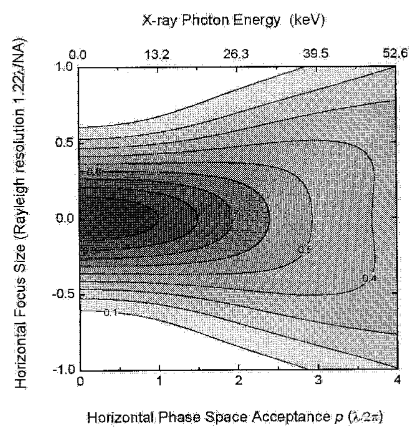

Figure 2. Focal spot size of a focusing optic versus the accepted phase space area in units of $\lambda/2\pi$. Given the fixed ERL emittance of 0.015 nm-rad, it shows that full undulator radiation within the central cone can be accepted up to 13 keV without significant degradation of resolution for SXM applications.

Based on the preliminary design parameters as listed in Table 1, the proposed high-coherence ERL source would be fully diffraction-limited up to 6.6 keV, and almost diffraction-limited up to 13 keV. As shown in Fig. 2, this implies that at these energies, full undulator radiation within the central cone can be utilized for SXM applications without significant degradation of spatial resolution determined by the optic, thus minimizing alignment problems from different components and increasing the x-ray flux to a specimen.

(2) *Phase-contrast imaging and microscopy*:

Phase-contrast microscopy has several forms [5]. The Zernike phase contrast method [19], based on phase shifting the zeroth order wave by $\pm\pi/2$ in transmission x-ray microscopy, does not require full coherence in the incident beam. On the other hand, high coherence is required for several other types of phase-contrast techniques, which include phase-contrast imaging in the near-field Fresnel regime, coherent scattering microscopy, and x-ray holographic applications.

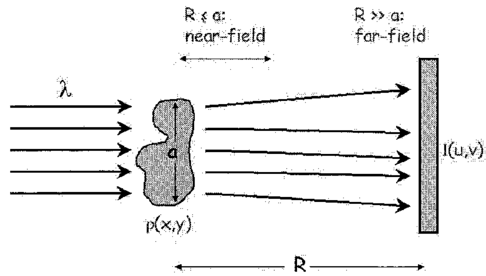

Figure3. Schematic illustration of phase-contrast imaging in the near-field or Fresnel diffraction, and the far-field or the Fraunhofer diffraction regimes.

Phase-contrast imaging in the Fresnel regime (Fig. 3), normally referred to simply as *phase-contrast imaging*, is a form of Gabor in-line holography [20-22]. It makes use of the interference between the direct beam and the refracted x-rays from a close-by region in the specimen. The degree of coherence required depends on the spatial frequency of interest, which is the inverse of average spacing between the inhomogeneities in the sample that one wants to study. For a given detector to sample distance R and x-ray wavelength λ, the incident x-ray beam should be transversely coherent over the size of the first Fresnel zone $\sqrt{\lambda R}$, and not necessarily over the whole specimen [23]. A higher degree of coherence would allow quantitative holographic phase reconstruction of a phase object. In this regard, the larger coherence width and much higher coherent flux from an ERL source would make phase-contrast imaging an attractive technique for time-resolved dynamic studies of structural features in alloys and other materials science specimens. It may also be possible to visualize phase imaging with x-ray microscopy, which would improve spatial resolution by projection [24].

(3) *Coherent diffraction x-ray microscopy*:

In phase-contrast imaging described above, the spatial resolution is in practice limited by the pixel size of a two-dimensional detector. To overcome this limit, one can move the detector further into the far-field or Fraunhofer diffraction regime to record a complete diffraction pattern from the specimen as illustrated in Fig. 4. When the specimen is illuminated by fully coherent radiation, the diffraction pattern is equivalent to a continuous Fourier transform intensity map from the specimen and can be inverted by advanced phasing methods to reconstruct the original object [7-11].

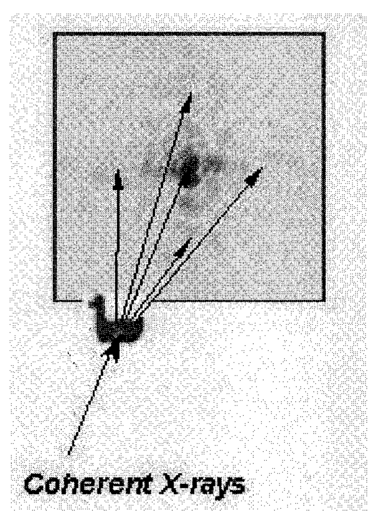

Figure 4. Coherent x-ray scattering from noncrystalline materials that yields a continuous Fourier transform intensity pattern, from which the object can be reconstructed by a iterative phasing technique.

There has been considerable scientific interest and research activity in recent years devoted to the topic of coherent scattering and phase reconstruction. The original idea was proposed by Sayre [25] in the early 1980's, but not until recently the principle of the so-called over-sampling or iterative phasing algorithm has been demonstrated by Miao *et al.* [7] in 1999 and by He *et al.* [10] in 2002. The same iterative phasing method has been used to reconstruct images of gold nanocrystallites by Robinson *et al.* [8]. Very recently an *Escherichia coli* bacteria real space image has been reconstructed to 30 nm resolution using the technique [11], showing the potential of single particle diffraction to study biological structures in high resolution without the need for crystallization.

In principle, the technique of coherent diffraction does not have any intrinsic resolution limit. In practice, it is only limited by the weak scattering signal at high angles and by the radiation damage of the specimen especially for biological samples. The proposed ERL source offers a factor of 10^3 increase in coherent flux density compared to present-day facilities, yet the overall flux density is similar to the existing sources. Therefore it would be an ideal x-ray source for further advances in the area of coherent diffraction microscopy from noncrystalline and nanocrystalline specimens.

(4) *X-ray holographic techniques*:

X-ray holographic imaging refers to imaging techniques that make deliberate use of a separate beam as the reference wave for interference pattern recording [26-29]. The reference wave can be a simple point source generated by a focusing optic [26, 29], or a complex wave produced by a nearby object with known structure [27, 28]. In either case, the specimen is placed in the vicinity of the reference source, within the transverse coherence width of the incident beam. These techniques therefore require a highly coherent x-ray source such as the proposed ERL. Although x-ray holography is in its early development stage, further research and advances can be expected in this exciting area of x-ray imaging if a highly coherent source becomes available.

4. CONCLUSIONS

In summary, we have briefly described the basic parameters and properties of the proposed energy recovery linac (ERL) synchrotron source at Cornell. This ultra-high brilliance source would offer substantial improvements in transverse coherence area, in transverse beam shape, in degree of coherence, and in coherent x-ray intensities for a specimen. We believe that these source properties would open up new research and application opportunities in x-ray imaging and microscopy. Potential advantages include full coherent illumination of wide-aperture focusing optics, dynamic studies using phase-contrast imaging and microscopy, coherent diffraction microscopy on noncrystalline specimens, and development of x-ray holographic techniques.

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