Focus on X-ray Beams with High Coherence

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EDITORIAL

X-ray beams with high coherence

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Abstract. This editorial serves as the preface to a special issue of \textit{New Journal of Physics}, which collects together solicited papers on a common subject, x-ray beams with high coherence. We summarize the issue’s content, and explain why there is so much current interest both in the sources themselves and in the applications to the study of the structure of matter and its fluctuations (both spontaneous and driven). As this collection demonstrates, the field brings together accelerator physics in the design of new sources, particle physics in the design of detectors, and chemical and materials scientists who make use of the coherent beams produced.

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1. Introduction

This Focus Issue of \textit{New Journal of Physics} is devoted to the new applications of the coherence of the x-ray beams that are produced by the latest synchrotron and free-electron laser sources. As such, the new field is technology driven, but many of the applications are extensions of the

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Figure 1. Peak brilliance of storage ring and FEL sources.

traditional field of optics to the shorter wavelength range. This allows access to the nanometre and even the atomic length scale in many cases. The papers included here can generally be identified with one of three topics, ‘sources’, ‘structure’ and ‘fluctuations’, although there is considerable overlap between these.

2. Sources

Much of the excitement concerning ‘coherence-based’ experiments stems from the unprecedented properties of free-electron laser (FEL) sources. These Linac-based machines can produce ultrashort photon pulses of well below 100 femtoseconds’ (fs) duration, while providing pulse intensities in excess of $10^{12}$ photons pulse$^{-1}$. The corresponding peak brilliance of such a source thus supersedes modern third-generation synchrotron sources by 10 orders of magnitude. This is illustrated in figure 1. The number of photons per mode (usually referred to as the degeneracy parameter $\delta$) can reach values of $10^9$ or higher, thus underlining the laser-like nature of FEL radiation.

The coherence properties of FEL radiation can be described within the framework of statistical optics, with the help of field correlation functions. As shown by Saldin et al [11] and Geloni et al [21], the spectrum of self-amplified spontaneous emission (SASE)-type FELs may contain up to hundreds of spikes indicative of the limited temporal coherence of these machines. The coherence times are typically about 1 fs. The degree of transverse coherence
Table 1. Wavelength and availability of FEL sources.

| FEL Source       | Wavelength (Å) | Start Operation |
|------------------|----------------|-----------------|
| FLASH (GER)      | 60 (45)        | 2005            |
| LCLS (USA)       | 1.5            | 2009            |
| FERMI (IT)       | 30             | 2010/2011       |
| SCSS (JP)        | 1              | 2011            |
| SPARX (IT)       | 15             | 2014            |
| European XFEL    | 1              | 2014/2015       |
| SWISSFEL (CH)    | 1              | 2016            |
| PAL (Korea)      | 1              | 2016            |

is generally high, with values close to unity. The high gain of the amplification process, and the exceptionally bright electron beams, give rise to the exceptional peak brilliance and the degeneracy parameter of x-ray FELs.

The first SASE-type FEL, in operation since 2005 in the vacuum ultraviolet (VUV) and soft x-ray regimes, is FLASH at DESY/Hamburg. An overview of coherence-based experiments carried out there is given by Treusch et al [12]. The linac coherent light source (LCLS) started user operation in 2009, producing for the first time 1.5 Å FEL radiation. In this issue, Boutet et al [19] describes the coherent x-ray imaging (CXI) instrument at the LCLS. Worldwide, there are quite a number of FEL projects in the planning, construction or commissioning phase, as summarized in table 1. Two FEL projects, the FERMI FEL at Elettra and the SwissFEL X-Ray Laser, and their related science programmes are described in more detail by Allaria et al [18] and Patterson et al, respectively.

Low repetition rate is a characteristic feature of FEL sources (120 Hz for LCLS or up to 30 000 pulses s\(^{-1}\) sorted into 10–30 macropulses for the European XFEL). This can be a disadvantage for certain classes of experiment, but will eventually be overcome by energy-recovery linac (ERL) machines. The Cornell project described by Bilderback et al [13] comprises a 5 GeV ERL with GHz repetition rates, typically 2 ps long pulses available simultaneously to all x-ray beamlines at the ERL. This will allow the whole portfolio of coherence-based techniques to be used.

3. Structure

Coherent x-ray diffraction (CXD) is the name given to the application of coherent x-ray beams when they are used to solve structure in the general sense. Traditionally, the field has been limited to crystal structure determination of the position of atoms within a unit cell, the basis of x-ray crystallography. The limitation to crystals was always attributed to the need to solve the ‘phase problem’, for which decades of progress has led to many elegant solutions.

Coherence offers a new general solution to the phase problem, which was first stated in a short paper by Sayre in 1952 [25], closely following the conclusions of Shannon in the field of communication theory [26]. Sayre pointed out that the continuous diffraction pattern from a non-crystallographic object was overdetermined, and so could be inverted to a structure. This result has been confirmed by subsequent mathematical analysis of the problem in a number of
different contexts. It was not well appreciated at the time that there was an implicit assumption of coherence behind these results. With hard x-rays, we had to wait until the turn of the 21st century for the first demonstration that the methods worked in practice [27]. The delay was partly due to the need to wait for the development of suitably bright x-ray sources to provide the necessary coherence.

Because CXD methods are distinguished from the crystallographic case by the need for a non-crystalline sample, there is a strong overlap with the fields of imaging, microscopy and holography. There is also a strong synergy with the current interest in nanotechnology. This comes about because nanoparticles are not really crystalline in the mathematical sense, but contain inherent strains associated with their facets, vertices and edges. Many of the useful applications of nanoparticles can be attributed to their structure in this way.

In this issue, there are three papers devoted to methodological developments of the experiments and algorithms needed to invert the diffraction patterns. There is a paper on Fresnel coherent diffractive imaging concerning the treatment and analysis of data by Williams et al [17], and one concerning the imaging of complex density in silver nanocubes by Harder et al [15]. A variation of a method called Ptychography [28] has been applied to coherent diffractive imaging of weakly scattering specimens by Dierolf et al [14]. Lastly, there is a paper on holographic and diffractive x-ray imaging using waveguides as quasi-point sources by Giewekemeyer et al [9].

There are several papers concerning the application of CXD methods to strain mapping. There is a general study of crystal strains using CXD with examples of zeolites by Cha et al [20]. There is also an analysis of strain and stacking faults in single nanowires using Bragg coherent diffraction imaging by Favre-Nicolin et al [10]; this paper illustrates the technological application of nanocrystals for the development of new semiconductor devices. Diaz et al [5] have carried out a feasibility study for imaging the displacement field within epitaxial nanostructures by coherent diffraction. Application to polycrystalline thin films is investigated by Vaxelaire et al [16] in a paper on the methodology for studying strain inhomogeneities during in situ thermal loading.

The appearance of x-ray free-electron lasers (XFELs) has led to exciting new developments such as the single-shot ‘differact and destroy’ approach that promises to prevent radiation damage. The structure of a single particle from scattering by many particles, randomly oriented about an axis, is discussed by Saldin et al [6] because it promises a structure solution without crystallization. Coherent imaging of biological samples with femtosecond pulses at the FEL FLASH is presented by Mancuso et al [2]. The dose requirements for resolving a given feature in an object by CXD imaging is then addressed by Schropp et al [8]. These methods have quite promising applications in biology, such as the mapping of the conformations of biological assemblies by Schwander et al [4] and the potential for 2D crystallography of membrane proteins at future xFEL sources, which is discussed by Kewish et al [1].

4. Fluctuations

It was realized before the application of coherence for determining structure that the intensity at each point in a speckled coherent diffraction pattern would change when the structure fluctuates. This allows a systematic measurement of the relaxation times of fluctuations as a function of their length scale. Photon correlation spectroscopy (PCS) [29] was well established using lasers.
for this purpose long before x-ray beams with sufficient coherence came along. The advantage of using x-rays, of course, is that shorter distances can be probed, possibly reaching the atomic length scale.

In this issue, there is a paper about microscopic return point memory in magnetic Co/Pd multilayer films by Seu et al [24], where the need to invert the data to get real space images is avoided using a clever trick. In their paper, Madsen et al [22] take us beyond the commonly used simple exponential correlation functions in the study of equilibrium dynamics to explain the stretched behaviour that is sometimes seen. Looking to the future, the paper by Fluerasu et al [23] describes some of the advances that have been made with the development of the x-ray PCS technique at the new National Synchrotron Light Source II, where a dedicated XPCS beamline is under construction. This machine has the second-highest coherent x-ray flux of all the synchrotron sources that are planned worldwide. XPCS with a steady state source is sensitive to spontaneous fluctuations arising from thermal excitation. The pulsed XFEL sources described above will also encourage the development of new techniques for the study of the susceptibility of matter through its impulse response, which provides the same information in a complementary way.

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