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Revisiting Bragg's X-ray microscope: Scatter based optical transient grating detection of pulsed ionising radiation

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ABSTRACT

Transient optical gratings for detecting ultrafast signals are routine for temporally resolved photochemical investigations. Many processes can contribute to the formation of such gratings; we indicate use of optically scattering centres that can be formed with highly variable latencies in different materials and devices using ionising radiation. Coherent light scattered by these centres can form the short-wavelength-to-optical-wavelength, incoherent-to-coherent basis of a Bragg X-ray microscope, with inherent scope for optical phasing. Depending on the dynamics of the medium chosen, the way is open to both ultrafast pulsed and integrating measurements. For experiments employing brief pulses, we discuss high-dynamic-range short-wavelength diffraction measurements with real-time optical reconstructions. Applications to optical real-time X-ray phase-retrieval are considered.

1. Introduction

Colour centres induced by ionising radiations are an aspect of conventional photography, for which many relevant chemistries were highly evolved by the time radioactivity and X-rays were discovered and explored using these detection techniques. Through the maturity of photographic emulsion technologies, they were prerequisites for successful crystallography structure solutions by 1913 [1,2]. Studies of formation, kinetics, diffusion and the many decay mechanisms of colour centres [3–8] also formed the background in which today's semiconductor radiation detectors were first developed [9–11]. Optical reconstruction of X-ray diffraction data enjoyed its first successes in 1929 [12]. This early colour centre work retains some relevance to our proposal, which aims at the optically scattering disturbances ionising radiation can produce in optically transparent media, and their potential in the broad context of modern incarnations of Bragg's optical X-ray microscope [13,14].

Today, brilliant X-ray sources, pulsed lasers and visible-light spatial light modulators (SLMs) are far better able to address the corresponding tasks of X-ray diffraction, optical readout and imposition of optical phase in optical reconstructions. In addition, computational techniques have been greatly refined for the task of phase determination. This motivates the vision of the present proposal: contemporary revisitations and variants of Bragg's X-ray microscope as a powerful and flexible research technique. Prior to these developments, an impressive indication of the state of the art of optical reconstruction of X-ray data may be found in Refs. [15–19]. Despite many subsequent developments in optical computing [20–22], a reason for the present latency of Bragg's X-ray microscope, which is equally applicable to other short-wavelength radiation scattering data [23], is the versatility of digital computation when coupled to semiconductor detectors [24]. Present widespread interest [25] and developments in distributable optically driven ultrafast X-ray generation [26–29] stand poised to re-awaken analogue optical reconstruction approaches in high-brilliance short-wavelength diffraction contexts. This is particularly true in ultrafast measurements, where optical computation permits reconstruction in experimental real time, while avoiding complications surrounding semiconductor detector pileup, pixelation and readout.

Timescales denoted as "ultrafast" in this work are in the context of contemporary pump–probe optical measurements, where detectors almost invariably operate in an integrating mode. A typical sequence of events leading to detection is: (1) a sample is pumped by a stimulus; (2) the effect of the stimulus is probed (in this case using short wavelength radiation); (3) radiation events are produced in a detector; and (4) the events are read out of the detector. Items 1 and 2 set the measurable timescale from the perspective of sample interrogation. Importantly they are decoupled from detection aspects occurring in items 3 and 4, which, in this work, are a subsequent pump–probe measurement. From a sample perspective, the term "ultrafast" can be used in whatever way items 1 and 2 permit. For example, sub-femtosecond measurements are a routine aspect of

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optical coherent control and 2D spectroscopy experiments [30,31], and it can be anticipated that they will become so in hard X-ray experiments [28]. From the perspective of items 3 and 4, which are the topic of this work, timescales vary enormously. Integrating modalities of Bragg's X-ray microscopy have been demonstrated using photographic film [23]. This work places greater emphasis on transient modalities, whose scope in contemporary studies [32] is clear. The vast ranges of timescales relevant to items 3 and 4, and obtainable in even just one example detection medium (water) [33–36], fully encompass those in conventional detectors such as charge-coupled devices (CCDs), complementary metal oxide silicon (CMOS) and more recent serial time encoded imaging technologies [37], any of which could be used for eventual optical detection in this work.

Fast and ultrafast synchronously pulsed X-ray/laser capabilities enable the study of transient molecular structures [38,39], a capability that is rapidly evolving on sub-picosecond timescales, in non-crystalline samples and on a laboratory scale [32,40–43]. Atomic spatial resolution is natural using correspondingly short X-rays or matter waves such as electrons or neutrons. Many innovative approaches have evolved for sub-wavelength optical steady-state microscopies [44–49]. Nevertheless, in fundamental chemistry contexts the usefulness of short wavelengths persists through the sample length scales accessible by the Ewald sphere [50], and the readily measurable scattering angles that result. On chemical dynamics timescales, short-wavelength approaches have no equal for the examination of atomic-resolution molecular structure.

Small-molecule and protein crystallographic databases demonstrate numerous successful phase reconstructions. The phase problem is common to most diffractive and temporal spectroscopy techniques [51,52]. Experimental arrangements are possible that provide less or more phase information, allowing increasingly non-iterative retrieval approaches (e.g. using masks, coded apertures or anomalous scattering in spatial studies, or, for ultrafast spectral examinations, heterodyne detection techniques) that in effect constitute holography in spatial [53] or temporal domains. The possibility of optical reconstruction is then well established [54–56]. Newer phase-retrieval approaches that could be incorporated into optical computations in the spirit of Bragg's X-ray microscope (e.g. using CCD/CMOS and SLM optical-computer interfaces) include the non-iterative methods of Nakajima [57] or Podorov et al. [58] for phase retrieval from far-field diffraction patterns, together with approaches based on transport-of-intensity concepts [59–61], ankylography [62] and the application of genetic algorithms [63,64] and optical neural networks to phase retrieval [65–67]. For spatial microscopy a wealth of optical filtering and computation capabilities has evolved [18,20–22], permitting a good deal of information processing to be carried out using analogue optical "computing" prior to any digital processing.

We will not further elaborate on the many viable approaches to short-wavelength phase retrieval. Instead, the key question for this work is how to interface pulsed monochromatic ionising radiation scattered by a sample to a coherent light field, i.e. the use of optical scatter from transient radiation-induced centres as the basis for detection in optically-reconstructing short-wavelength ionising radiation microscopes. Admittedly, a similar end has been achieved using permanent photographic techniques [23,68], and, in a transient guise, the essential incoherent-to-coherent, wavelength-to-wavelength interfacing has been achieved in fully optical contexts [69–71]. This work indicates the extension to time-resolved short wavelength studies. To our knowledge this is also the first suggestion of the use of SLMs to impose the optical phase where necessary in Bragg X-ray microscopy contexts, despite the now-widespread use of SLMs in commercial electronics [72].

Our proposal's novelty reflects widespread acceptance of electronic semiconductor detection and computational technologies as standard procedure for short-wavelength scatter and phase-reconstruction problems. This may have happened because optical technologies (especially lasers and SLMs) were insufficiently developed at a time when electronics and associated digital computation were enjoying remarkable advances. Computationally based analyses are readily adapted to non-linear problems, and represent both the original and present state of the art in short-wavelength diffraction-based reconstruction. It is hoped that the present work will reinvigorate the elegant early work on X-ray microscopes using all-optical reconstruction [18,19,54], now with vastly improved technological opportunities.

2. Structural dynamics and the problem of X-ray detector dynamic range

In pixelated semiconductor X-ray detectors, the measurable dynamic range can be restrictive [11,73]. One contributing aspect is that for non-holographic scatter techniques, only the squared modulus (the intensity) of the complex wavefield is registered by the detector, corresponding to measuring only the moduli of the sample's spatial Fourier components; phase information is lost. Next, ultrafast measurements from short-pulsed X-ray sources [42,43] cannot use time-of-flight techniques for energy-resolution, such as are used in neutron and electron scattering momentum transfer experiments [74–78]. Instead, scattered X-ray photons arrive essentially simultaneously at a detector, for which counter-based approaches are impossible. Event pileup means the inability to energy-resolve individual photons for the critical determination of momentum transfer (the fundamental entity that must be established for diffraction-based structure solution). Instead knowledge of momentum transfer can be preserved by the use of monochromatic photons, for which narrow collimated beams probe only a single, thin Ewald sphere [50] in individual shots. In this case, event pileup simply contributes intensity at the detector surface, up to the point where detectable dynamic range becomes a limitation. Limited dynamic range is problematic in pulsed experiments for which a stroboscopic mode is difficult or impossible, for example when samples are destroyed by one or a few shots, for example, in X-ray free electron lasers (XFELs). Here, semiconductor detectors such as direct-detection CCDs can prove inadequate owing to the limited charge capacity of individual pixels [10,11]. Detector saturation may be anticipated and measures taken to avoid it, but with little opportunity for optimisation. Hence in XFEL studies it is desirable to extend the single-shot dynamic range of the detector. This is one of the opportunities afforded by detection schemes proposed in this work. Another is the possibility to avoid effects associated with pixelation.

For time-resolved work in three spatial dimensions the thin monochromatic Ewald reflection sphere must have its effective reciprocal space volume extended, within the timescale of the radiation pulse, to encompass structure factors that the sample presents. Herein lies the power of time-resolved powder [79], diffuse scatter [80], and polychromatic Laue crystallography [42,81] in ultrafast contexts [40,42]. Non-collimated beams can achieve a similar outcome, corresponding, as with the use of polychromatic radiation, to the sacrifice of X-ray source brilliance in one or other degree of the source's Liouville phase space [82]. Nevertheless, this work revisits the use of brilliant monochromatic X-ray sources [83], which provide two-dimensional Ewald-sphere cuts of reciprocal space that can realise or anticipate ultrafast structure studies [38,84], and for which a lesser or greater degree of control may be associated with the stimulus [85]. Extrapolating successful crystallographic results [38] to single-molecule scattering proposals at XFELs [86], a level of optimism can be appreciated in terms of available photons and measurement times. For single molecule and nanoscale studies, X-ray scattering cross sections are small, motivating relatively soft X-rays [87]. Unfortunately the corresponding Ewald-sphere is also