TRANSIENT OPTICAL GRATINGS FOR SHORT PULSE, SHORT WAVELENGTH IONISING RADIATION STUDIES – OPPORTUNITIES AND APPROACHES*

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Abstract

Loss of short wavelength phase information occurs when event sizes exceed radiation wavelengths, making conventional holography impossible above a material-dependent quantum energy limit. Despite this, and prior to the invention of holography or lasers, Bragg's X-ray microscope [1] opened the door to optical computation in short-wavelength studies using spatially coherent visible light, including phase retrieval methods. This optical approach lost ground to semiconductor detection and digital computing in the 1960s. Since then, visible optics including spatial light modulators, array detectors and femtosecond lasers have become widely available, routinely allowing versatile and computer-interfaced imposition of optical phase, molecular coherent control, and detection. Today, free electron lasers (FELs) begin to offer opportunities for atomic resolution and ultrafast pump-probe studies. Thus we investigate an overlooked aspect of Bragg's X-ray microscope: the incoherent ionising radiation to coherent visible (IICV) spatial light modulator (SLM) conversion that is a necessary prerequisite for coherent optical computations. Some potential opportunities and approaches are outlined.

OPPORTUNITIES

X-rays are a form of light whose wavelengths match molecular interatomic spacings. This underpins not only the routine use of X-rays for molecular and small object structure determinations, but also the early realisation that propagation of suitably phased spatially coherent visible light fields can "undo" the X-ray scattering process to obtain greatly magnified optical reconstructions of the invisibly small objects that caused the X-ray scatter. This is the basis of Bragg's X-ray microscope [1], which is a viable approach to the observation of molecular structures [2-9] using X-rays and other short wavelength radiations [10]. The importance of introducing phase information in the optical reconstruction procedure was recognised at the outset by Bragg. In the case of scattering of optical wavelengths, this phase problem was later addressed in the invention of holography by Gabor [11]. However, the holographic preservation of phase requires that event sizes in the recording medium are small compared to the radiation's wavelength. This condition does not obtain at wavelengths corresponding to interatomic distances [12]. Having thus lost the short wavelength phase information, it is necessary to both determine and re-introduce it in any subsequent reconstruction. Early methods were devised for doing this optically [6, 7], which nowadays would be accomplished through the computer-interfaced combination of lasers, SLMs [13] and optical image transducer technologies [14-16]. However, digital X-ray detection and computation technologies had meanwhile matured to the point of universal use, so that Bragg's X-ray microscope lapsed into historical obscurity.

We now face the prospects of brilliant X-ray FEL (XFEL) sources. With lasers, computer-interfaced optical imaging capabilities and SLMs already in widespread use, it is timely to re-examine the case for Bragg's X-ray microscopy and related studies, such as X-ray photon correlation spectroscopy (XPCS) [17-19]. Techniques for IICV conversion are a missing link that impedes their contemporary redevelopment. We offer the following motivations for overcoming this impediment. Depending on the chosen approach, the dynamic range of X-ray detection in short-pulse experiments is potentially very dramatically improved. One can avoid issues related to detector pixelation. Effective and affordable lenses, beamsplitters and coherent optical gain media are highly developed for optical wavelengths, while corresponding devices at X-ray wavelengths are restrictive at best. Optical laser diffraction-based computation is the natural cousin of monochromatic X-ray diffraction, with the former now enjoying a mature yet extremely widespread and active state of development [20-22]. Optical approaches offer clear paths to new developments, such as optical artificial intelligence possibilities for X-ray phase retrieval [23-25], and 3-dimensional holography [26] to retrieve phase information through transport of intensity concepts [27]. The maturity of ultrafast coherent control at the molecular level [28-30] points to in-situ short wavelength molecular structure observations on the same molecular dynamics timescales [31], while ultrafast X ray – optical pump-probe studies would be an integral aspect of sample, detection, and data reduction technologies. The monochromatic approach implicit in this work, though limited in its potential for ultrafast work [31, 32], represents an extension of earlier successful optical pump, X-ray probe developments for observing transient molecular structures [33, 34]. In short, addressing X-ray to visible, incoherent to coherent conversion, particularly as transient gratings in modern ultrafast pump-probe contexts, opens a new door to the science of X-ray detection and analysis.

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APPROACHES

Figure 1 sketches an arrangement that performs the essential IICV SLM conversion. Variations may offer advantages [32]. Note the critical role of spatial coherence in the optical field. The use of light emitting phosphors or incoherent observations of metastable colour centres through various relaxation channels is thus essentially unrelated, though such studies can be both inspirational and relevant [35]. To accomplish transient grating IICV, several approaches are possible. The route chosen will depend on demands of particular applications. We indicate the following technologies:

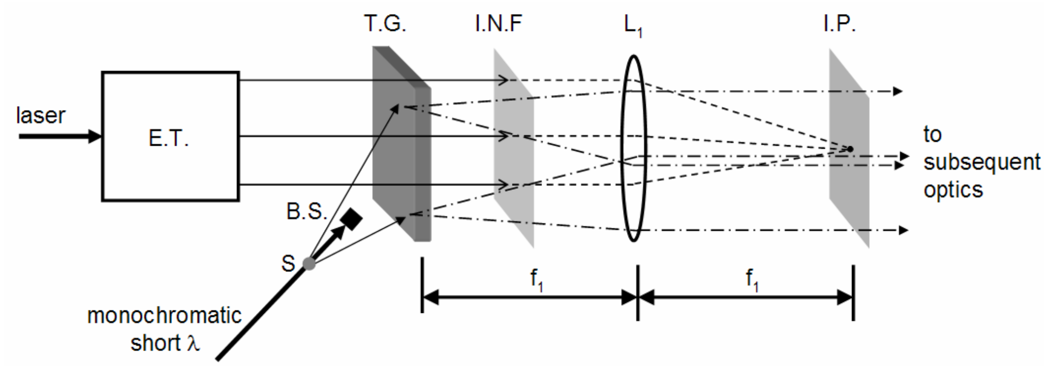

Figure 1: Pulsed monochromatic short wavelength radiation scatters from a sample S onto a transient grating window TG (BS = beamstop). Within the timescale of the transient grating, radiation-induced scattering centres in TG scatter an expanded and collimated pulsed laser beam (ET = expansion telescope). Unscattered laser light is prevented from further propagation by the (angularly selective) interference notch filter (INF) and/or spatial filtering at the image plane IP. Coherent optical processing can then be performed in subsequent optical stages.

Colour Centres Including Photography

Ionising radiation events can cause optically absorbing features in transparent media (or transparency in opaque media). Dye-based IICV SLMs have been developed in all-optical contexts [36] but apparently not for X-ray or other ionising radiation, despite widespread use of chemical dosimetry. Optical efficiency of absorption-based diffraction gratings is lower than for phase gratings [37, 38]. On the other hand, transient gratings involving F-centres and solvated electrons in condensed matter have been studied for well over a century, including large volumes of picosecond and femtosecond work in recent decades. They can offer extremely wide limits of temporal latency and exceptional dynamic range (being limited by recombination dynamics and the material's capacity to accommodate free electrons). Optical detectability of single X-ray events is questionable, but with the consolation that optical reconstructions are impossible based on small numbers of scattered ionising quanta (a large ensemble of events is essential, implying no need for their individual detectability). Chemical imaging procedures incorporating gain (typically photography) include radiation-induced polymerisation [39]. Gain improves sensitivity, but at the expense of available dynamic range and spatial resolution. It also implies a drop in chemical free energy, indicating that starting materials are in a metastable state and that changes are irreversible. The ability to rapidly regenerate the metastable state is then required if used as a transient optical grating. Regeneration could be by optical, electrical, pressure/acoustic, thermal or other means.

Liquid Crystal X-ray Light Value

In conjunction with optical polarisers, and for medical X-ray imaging, these devices have been under development for several years [40]. To our knowledge, they have not been developed for IICV purposes, though the technology appears suitable. Charges induced by ionising radiation in a semiconductor cause local alterations of birefringence in an adjacent thin film of liquid crystal, leading to modulations of optical phase in a probing light field. Foreseeable complications are that the dynamic range for the optical phase modulation is limited by the possible extent of molecular configuration change in the liquid crystal. In addition the kinetic rates of the molecular reorientation processes are slow (typically 0.01 - 0.1 s) when compared to repetition rates in many pump-probe experiments (e.g. kilohertz).

Ferroelectrics

In ferroelectrics optical birefringence is modulated by local electrical fields, including free charges generated by radiation-induced ionisation in the bulk of the material. The effect on a spatially coherent light field is similar to the liquid crystal devices, though here unconstrained by macromolecular reorientation. Thus, through the depth of a ferroelectric crystal, local optical phase shifts can occur to the extent that phase wrapping occurs. The kinetics of the atomic repositioning leading to birefringence are also very different, being dependent on the proximity to the Curie temperature [37]. An important work in the present IICV context is [41]. Acoustic translation and spatiotemporal manipulation of excitonic states in related devices [42] raises the possibility of a level of control comparable to that employed in the readout mechanism of conventional charge coupled devices (CCDs).

Cloud and Bubble Chambers

Here, optical detectability of single events is associated with gain in a supersaturated vapour or supercritical liquid, with nucleation points caused by ionising radiation. Liquid-gas phase boundaries scatter light in both cases. Many theoretical and experimental examinations of the rate and growth mechanisms of the droplets/bubbles have been made [43-45], including in devices driven by pressure waves asnd ultrasound [46]. The latter devices could be temporally phase locked to pulsed ionising radiation sources (e.g. FELs) and pulsed interrogation lasers to permit control of the optical scatter of growing droplets/bubbles by modulating the amplitude and temporal phase of the acoustic field. Bubble chambers offer short stopping lengths for ionising radiations, as motivated in Figure 1. On the other hand, relatively long stopping distances in the gaseous medium of cloud chambers opens the possibility of recovering short wavelength phase information as the radiation field propagates away from a scattering sample, through transport of intensity concepts [27]. Experimentally this would involve holographic examination of the cloud density. A potential complication is that, depending on their energy, ejected photoelectrons can have considerable ranges in gases, lowering measurement resolution. Indeed photoelectron trails are mainly responsible for the cloud's visibility, as seen in the beautiful and pioneering X-ray pump, optical probe work of Wilson [47].

MOLECULAR STRUCTURE-DYNAMICS

We put aside the ubiquitous phase reconstruction problem for a moment, while noting that the use of monochromatic ionising radiation is implicit in the schemes above. The observation of molecular and sample structure factors requires their presentation within the narrow Ewald reflection sphere (which, for the particular case of crystal diffraction, corresponds to meeting the Bragg reflection condition). For FEL radiation sources, the product of source size, beam divergence, spectral width and pulse duration is deliberately minimised, to maximise the brilliance and associated spatial and temporal coherence. The consequently dense and narrow phase space occupation corresponds to extremely thin Ewald spheres in reciprocal space for diffraction. Thus only a two dimensional slice of the sample's four-dimensional structure factor gives observable diffraction in any one radiation pulse (samples possess three spatial dimensions and evolve temporally in pump-probe studies). Mapping out this four dimensional situation is then a major undertaking, the more so if the sample is destroyed on a shot-to-shot basis (by either pump or probe radiations). In monochromatic synchrotron-based crystallographic contexts this difficulty has been successfully overcome [34] by making suitable compromises of sample and experimental constraints, including temporal resolution [33]. Similar problems now face FEL pump-probe molecular studies.

One solution is to use a source that is less brilliant by virtue of its broader spectral bandwidth, allowing a continuum of Ewald spheres to simultaneously probe the structure factors of the sample in each radiation shot. This is the philosophy behind successful ultrafast broadband Laue pump-probe crystallography [48, 49], as well as neutron Laue crystallography and essentially all neutron time-of-flight instruments (where approaches are motivated to make best use of available flux). Beam divergence can be exploited similarly. More generally, there are fundamental reasons to reduce the brilliance of the radiation source (without reducing the basic photon flux), to allow pulsed X-ray observation of structural dynamics in samples. In ultrafast pump-probe contexts, and in conjunction with new detector developments, recognition of this situation has for several years formed the rationale for powerful new laboratory-based ultrafast X-ray techniques using laser-generated X-rays [31, 50].

SUMMARY

The appeal and viability of Bragg's X-ray microscope remains, and modern implementations are foreseeable as powerful XFELs come online. Transient grating IICV SLM detectors would play a key role in such developments. They should also find use in XPCS studies – which may emerge as a unique strength of XFEL sources – to enable instantaneous optical readout of a detection plane in the interval between two ultrashort pulses of scattered radiation. Thus we believe these technologies can contribute substantially to contemporary developments within combined laser-FEL science [51].

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