Results in Physics 4 (2014) 113–116

Contents lists available at ScienceDirect

Results in Physics

journal homepage: www.journals.elsevier.com/results-in-physics

Generation of highly coherent extreme ultraviolet source and its application in diffraction imaging

Hoang Vu Le, Khuong Ba Dinh, Peter Hannaford, Lap Van Dao [*]

Centre for Quantum and Optical Science, Swinburne University of Technology, Melbourne, VIC 3122, Australia

ARTICLE INFO

Article history:
Received 27 May 2014
Accepted 22 July 2014
Available online 1 August 2014

Keywords:
High harmonic generation
Laser applications
Diffraction imaging

ABSTRACT

We report the generation of small bandwidth highly coherent extreme ultraviolet radiation and the application of this source in coherent diffractive transmission microscope. Using a focussed narrow-bandwidth high harmonic generation (HHG) source with wavelength around 30 nm we achieve a resolution of ~45 nm with a sample size down to 3 μm × 3 μm in a very short exposure time of <8 s. The experimental scheme is able for imaging sub-10 nm scale objects with an inexpensive table-top, commercially available femtosecond laser system.

Introduction

Novel imaging and labelling techniques in visible optical microscopy, where a resolution of down to 20 nm can be achieved in special cases [1–3], can be used to study dynamic processes in living cells and nanostructures. To obtain very high resolution illumination at short wavelengths, high resolution lenses are required. In visible light microscopes, lenses can be made from precision-shaped glass, while magnetic fields and zone plates are used for focusing electrons and X-rays, respectively. When focusing optics are applied as an objective in imaging, high-frequency information cannot be collected by the lens and is excluded in the reconstruction of the image, which limits the resolution, and lens imperfections cause aberrations in the image. Optics in the X-ray and Extreme Ultraviolet (XUV) regions are more difficult to produce and are less effective. To avoid the use of lenses coherent diffraction imaging (CDI), or "lens-less imaging" [4], has been developed.

Coherent diffraction imaging in its original concept has a plane-wave geometry and requires a high degree of spatial coherence of the light source. The principle of CDI is simple, and a similar technique has been used in X-ray crystallography, but the experimental realisation of CDI is much more difficult. In crystallography the weak signal diffracted from a unit cell of a large crystal is accumulated coherently and the periodic arrangement of the atoms acts like an amplifier so that detection of the Bragg peaks is straightforward. The diffraction pattern from a non-crystalline specimen or a nano-crystal has a much weaker intensity distribution which requires a very high flux source for illumination. The incident beam needs to be highly coherent and the coherence length is related to the over-sampling ratio of the diffraction pattern [5,6]. For high resolution the diffraction intensity at high-angle diffraction needs to be recorded and in order to obtain high-quality diffraction patterns the dynamic range and quantum efficiency of the detector need to be high. Iterative retrieval of the phase of the far-field diffraction pattern permits reconstruction of the diffracting object with a spatial resolution that is limited, in principle, only by the wavelength of the incident radiation and the properties of the detector.

At present the availability of a bright coherent source, especially an X-ray source which in many CDI experiments is provided by a synchrotron [7] or a free-electron laser [8], is a bottleneck in the development of X-ray diffraction microscopy. The "beam time" at free- electron laser or third-generation synchrotron facilities has limited the number of groups that are currently working on CDI techniques and their application. High harmonic generation (HHG) sources provide not only a small laboratory-scale table-top setup, but they also deliver radiation with a very high degree of spatial and temporal coherence [9,10]. While third-generation synchrotron sources can be used effectively for CDI [11], the incident light requires significant spectral and spatial filtering which decreases the photon flux substantially in order to achieve the necessary lateral and longitudinal coherence. X-ray free electron-laser sources [8] are partially coherent and have a degree of coherence (~0.8) significantly less than that of visible lasers. The high degree of spatial coherence of HHG radiation and the small-scale source offer an inexpensive approach, complementary to large facilities.

---

[*] Corresponding author.
E-mail address: dvlap@swin.edu.au (L.V. Dao).

http://dx.doi.org/10.1016/j.rinp.2014.07.008
2211-3797/

Using 30 nm-wavelength high-harmonic beams for CDI, a spatial resolution of 90 nm with relatively long integration times (80 min) [12] and single shot imaging with 125 nm spatial resolution [13] have been achieved. When a source at a wavelength of 13 nm is used a resolution of ∼25 nm (∼2λ) has been achieved with an expose time of 30 s [14]. A single harmonic can be selected by using optical elements such as narrow-bandwidth multilayer mirrors but a significant loss of total harmonic intensity is incurred. Previous work has shown that it is possible to use a well characterised large bandwidth harmonic source for reduction of exposure time in a CDI experiment with a spatial resolution of ∼150 nm [15]. Using a HHG source, the sample cannot be located close to the HHG source because the thin metal filter used to block the fundamental laser beam is very easily damaged by the fundamental laser. We have recently reported that a resolution of ∼100 nm for a periodic sample image could be achieved from a 300 s integration-time diffraction pattern obtained with a few high harmonic order sources around 30 nm and a sample size of ∼15 μm. The size of the HHG beam at the sample spot was usually >1 mm and therefore the effective photon flux for illumination of a micro-scale sample was also low. By using XUV focusing mirrors, a single-harmonic beam can be confined to a smaller area that is comparable to the size of the sample. As a result the total photon flux through the sample is increased and hence the exposure time needed to capture a full dynamic range diffraction pattern can be significantly reduced.

In this paper, we report the generation of a small bandwidth high order harmonic source and its application in diffraction image to achieve a spatial resolution of ∼45 nm. A short exposure time of ∼8 s to image a small size (3 μm × 3 μm) sample can be achieved by using a focusing mirror with a narrow-bandwidth HHG source around 30 nm. We discuss the condition, where the plane-wave field or additional phase terms need to be considered in the reconstruction procedure. This new experimental scheme is very promising for imaging sub-10 nm scale objects with a table-top source based on a small inexpensive femtosecond laser system. Our extreme ultraviolet diffraction image transmission microscope is driven by a table-top femtosecond laser (<2 mJ pulse energy, 805 nm wavelength, 1 kHz repetition rate, 30 fs pulse duration) which is used in many laboratories worldwide.

In the coherent diffractive imaging technique a coherent light source is used to illuminate an object and the intensity of the diffraction pattern of the object is recorded. Based on diffraction theory the diffraction field at the detector plane ($U(x_0,y_0)$) from an object ($U(x_1,y_1)$) where x and y are horizontal and vertical co-ordinates is given by [16]:

$$U(x_0,y_0)=\frac{-e^{-2\pi iz/\lambda}}{i\lambda z}e^{-2\pi i(x_0^2+y_0^2)/2\lambda z}\times\iint_{-\infty}^{+\infty}U(x_1,y_1)e^{2\pi i(x_0x_1+y_0y_1)/\lambda z}dx_1dy_1 \tag{1}$$

where z is the distance between the sample and the detector and λ is the wavelength of the source. When an object with a complex transmission function $T(x_1,y_1)$ is illuminated by a light field the object amplitude is found by multiplying the incident wave by $T(x_1,y_1)$

$$U(x_1,y_1)=E(x_1,y_1)T(x_1,y_1) \tag{2}$$

When a mirror is used to focus a plane-wave field with amplitude E_0 a field with quadratic phase and Gaussian intensity contribution are created in the transmitted field [17] at the object plane

$$U(x_1,y_1)=E_0e^{\left(-\frac{x_1^2+y_1^2}{4\sigma_I^2}\right)}e^{-i\alpha_f(x_1^2+y_1^2)}T(x_1,y_1), \tag{3}$$

where α_f is the phase factor which is dependent on the distance to the focus, σ_I is the width of the intensity distribution. For a plane field in conventional CDI, $U(x_1,y_1)=E_0T(x_1,y_1)$. The recorded intensity at the detector plane is $I(x_0,y_0)=U(x_0,y_0)\,U^*(x_0,y_0)$. For a focusing field the variation of intensity and phase cross sample plane need to be considered. The sample, $T(x_1,y_1)$, can be reconstructed from the intensity distribution of the diffraction pattern when the phase information is known. Because only the intensity of the diffraction pattern is recorded a Fourier-based iterative phase retrieval algorithm is used to recover the phase for reconstruction of the image [6,18]. In order to successfully reconstruct the image of the object the conventional CDI experiment requires that the object needs to be illuminated by a well characterised coherent plane wavefront and a reasonably narrow bandwidth light source ($\lambda/\Delta\lambda>1000$) [6]. To capture the far-field diffraction data using the Fraunhofer approximation, the distance between the detector and object plane (z) is large and the criterion $z\gg\frac{L^2}{\lambda}$ is satisfied, where L is the maximum dimension of the object. For an object of maximum dimension L, the requirement that light from both edges of the scattering object experiences a path difference of less than one longitudinal coherence length when scattered at an angle θ is $\sigma_S>L\sin\theta$, where σ_S is the coherence length of the source. The resolution Δ of the reconstruction via $\Delta=\lambda/2\sin\theta$ is only limited by the wavelength of the light source and the degree of high-angle diffraction data. Considering the Rayleigh criterion the theoretical resolution of the CDI can be estimated from $\delta=0.61\lambda/\mathrm{NA}$, where NA is the numerical aperture. In a successfully reconstructed image, the size of each image pixel is given by $r=\alpha\frac{z\lambda}{pN}$, where N is the number of pixels in an N × N CCD array, $\alpha=0.94$ (Sparrow criterion) and 1.22 (Rayleigh criterion). For a given wavelength of the illuminating radiation, the resolution is linearly proportional to the distance z between the sample and the CCD and limited by the size of CCD pixels.

The harmonic generation was driven by a 1 kHz, 800 nm multi-pass multi-stage chirped-pulse amplifier laser system, which produces 30 fs pulses with an energy ∼2 mJ. The diagram of experimental setup is shown in Fig. 1. The detailed experimental configuration and the optimisation of the HHG are described elsewhere [10]. We have previously verified the extreme-ultraviolet emission scales quadratically with gas pressure and interaction length [19], demonstrating that the target is operating in the phase-matched regime. In order to block the fundamental driving laser and also its scattering into the detector the high harmonic beam passes through several pinholes with diameter <2 mm and a 200 nm-thick aluminium filter which has a 60% transmission in the wavelength range 17–60 nm. The XUV radiation is generated around the end of a long gas cell which has a glass window at the entrance and a ∼100 μm laser-drilled pinhole at the exit. By an appropriate choice of species of gas, gas pressure, interaction geometry, position of the laser focus and intensity and diameter of the laser beam, the harmonic emission can be phase-matched and confined to just a few harmonic orders [19]. A typical HHG spectrum is shown in Fig. 1a when argon gas is used as the active medium.

The good beam quality in the far-field of the harmonic and the small bandwidth of the harmonic spectrum indicate that phase matching is mainly satisfied along the propagation axis of the pump pulse [19]. The band-width of the 27th harmonic (around 30 nm) is small and we can obtain $\lambda/\Delta\lambda>250$. The coherence degree of the source is ∼0.99, which can be measured in a combination of a Young double slit and a spectrometer.

The HHG radiation is reflected by an XUV plane mirror and a 20 cm-radius focusing mirror at a reflection angle of ∼10° in a Z-configuration to obtain a good focusing spot. One strong narrow bandwidth harmonic at a wavelength around 30 nm (the 27th harmonic at 29.8 nm) is selected to illuminate the sample (see inset of Fig. 1b). Small contributions from the 25th and 29th