Ultramicroscopy 167 (2016) 1–4

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

X-ray holography with an atomic scatterer

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ARTICLE INFO

Article history:
Received 8 June 2015
Received in revised form
7 April 2016
Accepted 26 April 2016
Available online 27 April 2016

Keywords:
X-ray holography
Object reconstruction
Atomic resolution
Alignment

ABSTRACT

X-ray holography scheme with reference scatterer consisting of heavy atom as reference center and its link to an object consisting of several light atoms and using controlled variation of the alignment is represented. The scheme can reproduce an object in three dimensions with atomic resolution. The distorting factors of reconstruction are considered.

1. Introduction

In spite of significant progress in spatial resolution in electron and X-ray microscopy for crystals in case of disordered objects the achievement of atomic resolution still remains a challenging problem. Partial solution in case of electron microscopy consists in use of the aberration correction of electron lenses. An adequate lenses for X-ray beams do not exist. An alternative approach is a lensless imaging technique based on observation of diffraction and especially in holography mode. Significant limitation here is the required beam fluence. For coherent imaging an exposure determines the achievable resolution, and radiation damage sets the maximum dose. Promising solution of these problems can be in using novel ultrafast laser technique [1]. The results of the work [2] show that the degradation of the diffraction pattern from ultrafast electronic damage happens at fluences of at least one order of magnitude larger than provided at currently available free electrons laser (XFEL) sources. To date the resolution close to atomic was gained with X-ray beam of XFEL in observation of diffraction [3].

Diffraction pattern gives directly the autocorrelation function of an object while an image reconstruction can be complicated [4]. Conventional approaches to solving the phase problem in X-ray crystallography are Multiple Isomorphous Replacement (MIR), Single-wavelength anomalous diffraction (SAD) and Multi-wavelength anomalous dispersion (MAD) [5,6]. More straightforward reconstruction is admitted by holography scheme, especially in off-axis mode. An important factor in holography is the properties of the reference wave. It is well known that if the reference wave is "well-characterized", it can be build an efficient procedure of extracting the object from the diffraction pattern [7–10]. However, the main practical problem is precisely how to get such a "well-characterized" reference wave. In this work it is considered the reference wave arising in X-ray scattering by almost a dot atomic scatterer. If the reference and the object waves are formed by scattering of the incident wave of appropriate wavelength, then the spatial resolution is limited by the size of the reference scatterer.

Single atom is the natural choice as the reference scatterer for atomic resolution. This possibility for achieving atomic resolution in electron microscopy was developed in [11–14], where single atom or a row of atoms in crystal was called an atomic focuser. The last one configuration keeps atomic resolution in the plane normal to the row axis. Its use for in-line electron holography was studied in [15]. The possibility to use atomic scale objects as lens systems for corpuscular optics and holography was considered in [16,17]. In case of X-rays an interesting scheme using single atom as the detector in backscatter mode was developed in so called X-ray fluorescence holography [18,19]. The use of nanoparticle as reference scatterer proposed in work [20] to solve the problem of intensity will lead to the loss of resolution at the level of nanoparticle size.

The holography experiment with atomic scatterer can be realized by the scheme used in [3]. The practical problem is the fabrication of a sample with attached atomic scatterer. One possible way to make link with heavy atom is to attach molecule of haloalkanes, for example 1-iododecane. Such arrangement has additional advantage for alignment in external fields in the scheme with controlled variation of the alignment direction in space.

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http://dx.doi.org/10.1016/j.ultramic.2016.04.008
0304-3991/

In this paper we consider the feasibility of using an atomic scatterer as the reference wave source for X-ray holography and in particular requirements for relative intensities of the scattered waves. The most simple scheme is off-axis holography with reconstruction of two-dimensional projected scattering potential. This scheme needs no variation of the alignment angle. However more interesting is the scheme with reconstruction of scattering potential in three dimensions. We consider one of the possible variants of such scheme in next section.

2. Reconstruction of the autocorrelation function of scattering potential in three dimensions

For X-ray beam the scatterer can be described by means of the value

$$V(\mathbf{r}) = r_e\, \rho(\mathbf{r}),$$

where r_e is classical electron radius and $\rho(\mathbf{r})$ is electron density distribution. We refer to this value as scattering "potential" in analogy with electron optics.

Diffraction patens detected at large distance r_0 from the scatterer can be written as

$$H = |\Phi|^2 \sigma,$$

where $\sigma = |A|^2$ is scattering differential cross section and A is scattering amplitude which in first Born approximation can be written as

$$A(\mathbf{q}) = \int \exp(i\,\mathbf{q}\cdot\mathbf{r})\, V(\mathbf{r})\, d\mathbf{r},$$

where $\mathbf{q} = \mathbf{k} - \mathbf{k}'$, $\mathbf{k}'$ is the wave vector of the incident wave normal to the detector plane and $\mathbf{k}$ is the wave vector of the scattered wave and for elastic scattering they have equal modulus $k = k'$. The scattering amplitude has the form of Fourier transform of scattering potential

$$A(\mathbf{q}) = F(V)(\mathbf{q}).$$

So inverse Fourier transform of scattering cross section is related with autocorrelation function of scattering potential by conventional formula

$$IF(\sigma) = V \otimes V.$$

Note that we ignore the coefficients in Fourier transform since we need no absolute value of diffraction paten.

The prefactor Φ has the form

$$\Phi(\mathbf{n}) = \frac{\exp(i\,k\,r_0)}{r_0}(\mathbf{n}\times(\mathbf{n}\times\mathbf{E})),$$

where $\times$ is cross product, $\mathbf{n} = \mathbf{k}/k$ is the unit vector in direction of the scattered wave vector $\mathbf{k}$ and $\mathbf{E}$ is the vector of electric field strength.

Let us consider rotated potential described by rotation operator with rotation axis containing chosen coordinate center and parallel to the detector plane. Rotation operator with rotation angle φ is denoted as $\Omega(\varphi)$ and corresponding rotated potential is

$$V(\Omega(\varphi)\cdot\mathbf{r}).$$

Since rotation is orthogonal operator $\Omega(\varphi)^{-1} = \Omega(\varphi)^T$ a scattering amplitude for rotated potential can be written as

$$A(\Omega(\varphi)\cdot\mathbf{q}).$$

In the space of wave vectors consider a plane P normal to the vector of the incident wave $\mathbf{k}'$. It can be treated as dual to detector plane. Suppose we have some function $W(\boldsymbol{\kappa})$ of arbitrary wave vector $\boldsymbol{\kappa} \in R^3$, defined piecewise on the set of rotated planes $P(\varphi) = \{\boldsymbol{\kappa} = \Omega(\varphi)\cdot\mathbf{p} | \mathbf{p} \in P\}$.

Inverse Fourier transform of this function can be expressed in form

$$IF(W)(\mathbf{r}) = \int \exp(-i\,\boldsymbol{\kappa}\cdot\mathbf{r})\, W(\boldsymbol{\kappa})\; d\boldsymbol{\kappa} \quad =$$

$$\int_{\mathbf{p}\in P \quad \varphi\in[0,\ \pi)} \exp(-i\ \ (\Omega(\varphi)\cdot\mathbf{p})\cdot\mathbf{r})\ \ W(\Omega(\varphi)\cdot\mathbf{p})\ \left|\det\left(\frac{\partial\boldsymbol{\kappa}}{\partial(\mathbf{p},\varphi)}\right)\right|\ d\mathbf{p}\ d\varphi$$

Suppose that the function W is truncated on a sphere of radius K in the space of wave vectors. This gives the convolution of inverse Fourier transform with truncation function's Fourier transform $\Gamma(r) = K^3\ \gamma(Kr)$, $\gamma(a) = \frac{4\pi}{a^3}(\sin(a) - a\ \cos(a))$. A disk of radius K on the plane P with $p < K$ corresponds to a sphere of radius K in the space of wave vectors $\kappa < K$ and the convolution can be written as

$$(IF(W)*\Gamma)(\mathbf{r}) = \int_{\kappa<K} \exp(-i\,\boldsymbol{\kappa}\cdot\mathbf{r})\, W(\boldsymbol{\kappa})\; d\boldsymbol{\kappa} \quad =$$

$$\int_{p<K \quad \varphi\in[0,\ \pi)} \exp(-i\ \ (\Omega(\varphi)\cdot\mathbf{p})\cdot\mathbf{r})\ \ W(\Omega(\varphi)\cdot\mathbf{p})\ \left|\det\left(\frac{\partial\boldsymbol{\kappa}}{\partial(\mathbf{p},\varphi)}\right)\right|\ d\mathbf{p}\ d\varphi$$

This leads to smoothed inverse Fourier transform with spatial resolution about $\Delta \sim 2\pi/K$.

With detector aperture half angle α we have $K = k'\alpha$ and spatial resolution can be written in form $\Delta \sim \lambda/\alpha$, where $\lambda = \frac{2\pi}{k'}$ is wavelength. For given detector aperture angle a desirable resolution can be achieved with appropriate wavelength. At small aperture angles $|\alpha| << \pi$ following approximation holds $\mathbf{q} \cong \mathbf{p}$ and hence smoothed autocorrelation function of scattering potential can be reconstructed by the set of holograms of rotated object, assuming $W(\Omega(\varphi)\cdot\mathbf{p}) \cong H(\Omega(\varphi)\cdot\mathbf{q})$.

Note that for considered aperture angles the prefactor is practically constant $\Phi(\mathbf{n}) \approx \Phi(\mathbf{n}')$, $\mathbf{n}' = \mathbf{k}'/k'$, so that diffraction paten is given by the scattering cross section.

Another source of smoothing is the spread of alignment angles $\delta\varphi$. This factor can be estimated as $\Delta \sim l\cdot\delta\varphi$, where l is the scatterer size, and leads to estimation of required alignment strength at given resolution level $\delta\varphi \sim \Delta/l$.

Further we consider the case of sufficiently small spatial resolution Δ and neglect smoothing effects.

3. Distortions in reconstruction of the object

In this section we consider the factors affecting reconstruction of the object from the hologram. We write some conventional basic formulas related to holography principles in the form convenient for our presentation. According to holography principle scheme let us write total scattering potential as the sum of object and reference contributions

$$V(\mathbf{r}) = O(\mathbf{r}) + R(\mathbf{r}).$$

Autocorrelation function of scattering potential is

$$V \otimes V = O \otimes O + R \otimes O + O \otimes R + R \otimes R \tag{1}$$

For further analysis we consider reference scatterer consisting of heavy atom (reference center) and its link to an object consisting of several light atoms. The arrangement of the object and the reference scatterer is shown in Fig. 1. For the simpler estimates we use a zero-radius-potential approximation for atomic potentials, which has proved to be useful in the qualitative consideration of the problems in atomic physics [21]. Let us write reference potential as the sum of link potential L and heavy atom potential

$$R(\mathbf{r}) = L(\mathbf{r}) + Z\ \delta(\mathbf{r}), L(\mathbf{r}) = \sum_j Z_j\ \delta(\mathbf{r} - \mathbf{r}_j)$$

with effective nuclear charges $Z_j << Z$. Heavy atom center is chosen as natural coordinate center.