Instrumental distortion effects in atomic resolution neutron holography

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Abstract

The influence of instrumental effects has to be taken into account in assessing various limitations to the quality of holographic images obtained with thermal neutrons. In comparison with the traditional treatment of resolution effects in the field of neutron scattering this requires a somewhat different approach. The reconstruction of the position of a nucleus from a hologram leads to a spot-like distribution of finite width in real space and the knowledge of the functional shape of this distribution is of crucial importance. In this context, the choice of the angular range covered by the recording process, the wavelength as well as the wavelength spread of the neutrons have to be considered. In planning a particular experiment, a proper choice for each of these quantities has to be worked out. Instrumental resolution also defines a limiting distance in real space up to which neighbouring object nuclei can be distinguished. The mathematical background of the above considerations is discussed.

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1. Introduction

The basic concepts of atomic resolution holography are well known [1]. There are basically two approaches to realization. One is the so-called inside source method in which particular atoms embedded in the sample are used as a secondary source of spherical waves. For neutron holography the proton (i.e. the nucleus of the hydrogen atom) is the best source for this technique [2,3]. In the inside detector method, one uses particular atoms as detectors embedded in the sample. Atoms are suitable for this purpose if an absorption process is followed by prompt emission of photons or electrons which can be counted by a detector. In neutron holography, e.g. Cd or Gd nuclei may be used as detectors [4]. The mathematical expressions describing the holographic imaging process for neutron, X-ray and electron holography in a first approximation are similar both for the inside source and the inside detector technique. The detector or the source atom are taken as the origin in the respective holographic image. For the sake of simplicity, we restrict the discussion of inside source neutron holography to the case of monocrystalline samples though only long range orientational order would be required. Dealing with a monocrystal containing suitable source atoms and neglecting any instrumental effects as well as the presence of Bragg peaks the normalized intensity observed in a direction defined by \( \mathbf{k} \) is [1]:

\[
\frac{I(\mathbf{k})}{I_0} = \frac{1}{4\pi} \left| 1 + \sum_j \frac{f_j(\mathbf{k})}{R_j} e^{i(kR_j - kR)} \right|^2, \tag{1}
\]

\[
\frac{I(\mathbf{k})}{I_0} = \frac{1}{4\pi} + \frac{H(\mathbf{k})}{4\pi} + O \left( \frac{1}{R_j^2} \right), \tag{2}
\]

where \( H(\mathbf{k}) \) is the hologram in momentum space, \( \mathbf{k} \) is the wave-vector of the scattered wave, \( f(\mathbf{k}) \) and \( R_j \) stand for the scattering amplitude and the position of the \( j \)-th atom, respectively. The third term is negligible because the
interatomic distances are much higher than the scattering length of the atoms. In the case of neutron scattering, the scattering amplitude is given by the scattering length which is independent of $s$:

$$H(k) = \sum_j b_j \cos(kR_j - kR). \quad (3)$$

The reconstructed image at the position $r$ is

$$U(r) = \iiint H(k)e^{-i(kr - kr)} d^3k. \quad (4)$$

Commonly the solution of Eq. (4) can be expressed as

$$U(r) = \sum_j b_j \delta(r - R). \quad (5)$$

2. Instrumental effects

In a real experiment the recording of a hologram usually covers only a limited domain of a closed spherical surface in 3-dimensional $k$-space see e.g. paper [4]. In X-ray holography, the effects resulting from such restricted measurement domains were investigated by Marchesini [5]. In the present work, we extend this discussion to neutron holography including the effects of instrumental resolution. As a rule, presently available detectors scan the holographic modulation over a finite set of discrete points on a spherical surface around the origin. This means that the ideal hologram has to be multiplied by a $d$(detector)-function composed from a set of Dirac-delta functions corresponding to all measured points in $k$-space.

$$d(k) = \sum_i \delta(k - k_i). \quad (6)$$

The finite wavelength-distribution and collimation of the primary beam, the finite active size of the detector and the finite mosaicity of the sample cause distortions of the hologram. These distortions can be described applying an appropriate $g$(instrument)-function. As a consequence, the measured hologram is the convolution of the ideal hologram and of the instrument function. The $g$-function in Gaussian approximation:

$$g(k) = e^{-|k|^2/2\sigma^2}. \quad (7)$$

Then, the measured hologram ($H^{\exp}$) is

$$H^{\exp} = (H' * g) \cdot d, \quad (8)$$

$H'$ is the theoretical hologram defined above, $*$ denotes the operation of convolution. Applying Fourier transformation in Eq. (8) the convolution of two functions changes into the multiplication of their Fourier transform:

$$U^{\exp} = (U' \cdot G) * D. \quad (9)$$

Here $U^{\exp}$ and $U'$ is the Fourier transform of $H^{\exp}$ and $H'$, respectively, $G$ is the Fourier transform of the $g$(instrument)-function:

$$G(R) = e^{-\left(r^2/2\sigma^2\right)} R^2. \quad (10)$$

$D$ stands for the Fourier transform of the $d$(detector)-function. Using Eqs. (9) and (10), the measured hologram has the form

$$U^{\exp}(r) = [U'(r)e^{-(r^2/2\sigma^2)}] * D. \quad (11)$$

In monochromatic case, the Fourier transformation and the reconstruction defined in Eq. (4) differ only in an $e^{ikr}$ phase factor. By investigating Eq. (11), one can arrive at some interesting result: the instrumental resolution and the imperfectness of the sample (i.e. the mosaicity, the thermal motion of the atoms) determine the size of the observable space but do not determine the size of the atomic peak in the reconstructed image. However, the size and the shape of the atomic peak is defined by the $D$ function, which inherits the symmetry of the measured domain and whose amplitude decreases as $r^{-1}$. This slow decrease causes overlapping of the atomic peaks and gives rise to some shift of the maxima of the peaks. Using a wider instrument function in the measurement, the number of the visible atoms is decreasing and the shifts caused by overlapping will be smaller. It is important to note, that the numerical convolution after the measurement decreases the size of the visible space but does not influence the shifts.

3. Model calculations

In order to illustrate the above mentioned effects, we carried out model calculations for various situations. For the sake of simplicity, we dealt with the holographic imaging of a set of points arranged on a primitive cubic lattice with a lattice parameter of 1 Å. The hologram was simulated in two ways. First, the hologram was calculated in 3D $k$-space. Second, the same object was imaged over a spherical surface with radius of $8\pi$ Å$^{-1}$ in $k$-space. The most distant atom is at a distance of 5 Å from the origin. Fig. 1 well displays that when the imaging is limited to the
spherical surface, the reconstructed hologram contains a lot of side oscillations making the image less clear and shifting the atomic peaks. These oscillations originate in the holographic image of one single atom: $\sin(kr)/kr$ which is the Fourier transform of the spherical surface. As a next step, we were trying to illustrate the effect of the restricted angular area of the data collection. Instead of $4\pi$ solid angle only an equatorial belt-shaped area was considered, i.e. the angle $\theta$ varied between 0 and $\pi/4$ while $\phi$ varied over the full range $2\pi$. For the sake of simplicity, we considered only the first neighbours (6 atoms) for modelling at $k = 4\pi \text{Å}^{-1}$. The result presented in Fig. 2 shows that this restriction of the angular area gives rise to significant differences in the images of the nuclei obtained for the different angular directions. These differences are caused by the asymmetrical measurement domain (detector function). This asymmetry causes different shifts of the position of the atomic peaks in different directions. Finally, the effect of the finite width of the instrument function is demonstrated (see Fig. 3). This effect is reflected in the suppression of the amplitude of remote nuclei in the reconstructed holographic image. (Remarks for the figures: each figure shows the amplitude of the complex reconstructed image along a straight line defined by the source nucleus and one of the first neighbours. The origin is at the position of the source nucleus, $R$ is the distance from the source, the vertical bars mark the real atomic positions. The ordinates in all figures are normalized to the highest peak.)

4. Results and discussions

The finite measurement domain causes peak broadening and side oscillations in the reconstructed hologram. The sum of oscillations could give rise to false peaks and shift the position of real atomic peaks. The finite resolution cause suppressing of the intensity of the theoretical reconstructed image. Worsening the resolution causes not only decreasing the intensity of peaks of the farther atoms but decreasing its side oscillations too. To quantify the shifts of the atomic peaks, the size of the visible space, and discern the real and false peaks model calculations should be made.

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References