# New Developments in Atomic Resolution Holography

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## Abstract

Atomic resolution neutron holography (ARNH) constitutes a novel technique to obtain structural information in three dimensions of the local crystal structure. It is based on the recording of the interference of neutron waves coherently scattered by nuclei located on a crystal lattice with a suitable reference wave. In the present talk we discuss the ways of carrying out successful observation of ARNH. Amongst others the optimization of the instrumentation and the estimation of the signal-to-noise ratio are considered. At present time the ARNH is demonstrated on more than five systems. The involvement of an absolutely new approach, the so called double reconstruction method, improves significantly the observation of the holographic signal allowing to suppress the side oscillation inherently appearing in the holographic intensity distribution. A practical application of the ARNH is demonstrated. Also some unresolved problematic observation is presented for discussion.

## 1. Introduction

Matter wave postulated by de-Broglie allows using neutrons for carrying out holographic experiments. There are two ways of realization. Either the neutron waves are emitted from a point-like source (internal source concept), or are detected by a point-like detector embedded in the sample made of a single crystal (internal detector concept).

The first wave that reaches the detector directly serves as the reference wave; the second one is scattered by the object of interest and subsequently interferes with the reference wave (See Figure 1.)

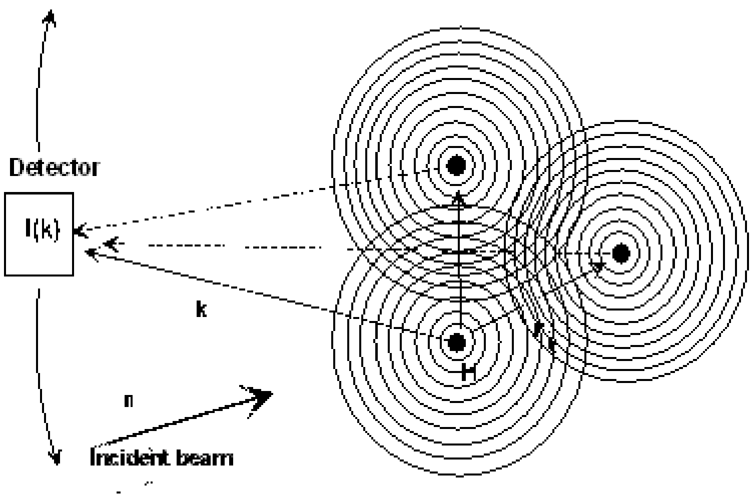


*Figure 1. The internal source concept*

The Figure 2. shows the schematic view of the internal detector concept.

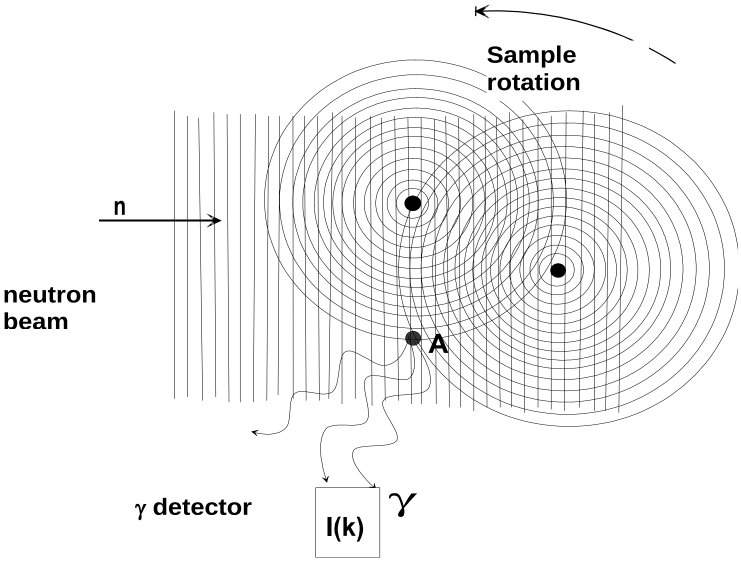


*Figure 2. The scheme of the internal detector concept*

In order to realize the internal source concept the huge incoherent scattering of the hydrogen nucleus was used because neutrons incoherently scattered by hydrogen nuclei form spherical waves. The point-like detector nucleus should possess a high cross-section for neutron capture and the absorption process should trigger a prompt nuclear reaction, generating a γ-ray cascade. This condition is satisfied by several nuclei as $^{113}Cd$, $^{10}B$, $^{149}Sm$ etc. More details are given in [1].

## Mathematical description

$$I(\mathbf{k}) = \frac{I_0}{4\pi R^2}\left|1+\sum_j a_j(\mathbf{k})\right|^2 = \frac{I_0}{4\pi R^2}\left[1+2\Re\left(\sum_j a_j(\mathbf{k})\right)+\left|\sum_j a_j(\mathbf{k})\right|^2\right] \qquad (1)$$

Reference beam — Object beam — Background — Hologram — Negligible

$$I(\mathbf{k}) = \frac{I_0}{4\pi R^2}\left(1+\sum_j \chi_j(\mathbf{k})\right) = \frac{I_0}{4\pi R^2}\left(1+\chi(\mathbf{k})\right) \qquad (2)$$

The function $\chi(\mathbf{k})$ stands for the holographic term which in more explicit way is expressed as

$$\chi(\mathbf{k}) = \sum_{j=1}^{n} \frac{-2b_j}{r_j} \cos\left(kr_j - \mathbf{k}\mathbf{r}_j\right)$$

Here $b_j$ stands for the scattering length. For getting the holographic image in the real space the Barton's procedure (the Helmholtz-Kirchhoff transformation) [2] has to be applied, i.e.

$$U(\bar{R}) = \frac{1}{V_{M\,n}} \int_{M_n} \chi(\bar{k}) \exp\left(i\left(\bar{k}\bar{R} - kR\right)\right) d^n\bar{k}$$

Here, $U(\bar{R})$ is the complex amplitude of the reconstructed holographic image at the point $\bar{R}$ in real space, $M_n$ is the $n$-dimensional domain of the measurement in $\bar{k}$ -space, where $n = 3$ in real three-dimensional measurement, i.e. time-of flight neutron holography, and $n = 2$ in other cases, and $V_M$ is the $n$-dimensional volume of $M_n$. If $n = 2$, $M_n$ is part of the surface of a sphere with a radius $2\pi/\lambda$, and $d^n\bar{k}$ means the elementary surface on the sphere.

The intensity of the holographic image is

$$I(\mathbf{R}) = \left|U(\mathbf{R})\right|^2$$

For two nuclei $n$ and $m$ occupying symmetrical positions ($\mathbf{r}_n = -\mathbf{r}_m$), this relation allows rewriting the holographic term as a sum over separate symmetrical pairs in the form

$$\chi(\mathbf{k}) = 2\sum_{n} \frac{b_n}{r_n} \left[\cos\left(kr_n - \mathbf{k}\mathbf{r}_n\right) + \cos\left(kr_n + \mathbf{k}\mathbf{r}_n\right)\right]$$

where the summation now is made over symmetrical atomic pairs. Using a simple trigonometric transformation one obtains

$$\chi(\mathbf{k}) = 4\sum_{i} \frac{b_n}{r_n} \cos\left(kr_n\right) \cos\left(\mathbf{k}\mathbf{r}_n\right)$$

Now, applying the Helmholtz-Kirchhoff transformation

$$U(\mathbf{R}) = \int_{\sigma_{\bar{k}}} \chi(\mathbf{k}) \exp\left(i\mathbf{k}\mathbf{r}_n\right) d\sigma_{\bar{k}} = 4\sum_{n} \frac{b_n}{r_n} \cos\left(kr_n\right) \int_{\sigma_{\bar{k}}} \cos\left(\mathbf{k}\mathbf{r}_n\right) \exp\left(i\mathbf{k}\mathbf{r}\right) d\sigma_{\bar{k}}$$

The observed intensity of the reconstructed hologram is obtained from the squared absolute value of $U(\mathbf{R})$. Then the holographic term gets separated into the product of two contributions. The first one reflects the modulation of the maximum value of the intensity proportionally to

$$I(\mathbf{R}) = \left|U(\mathbf{R})\right|^2 \sim \left[\frac{\cos\left(\mathrm{kr_n}\right)}{\mathrm{r_n}}\right]^2$$

This relationship is called as the cosines rule.

## Remark on the statistical noise

The statistical noise superimposed on a hologram during the recording procedure will, in turn, affect the value of noise in the reconstructed image in real space. By using properly the statistical error propagation [3] it was proven that the level of the statistical noise in the reconstructed hologram is proportional to the square root of the total number of counts collected implying that a successful holographic reconstruction is possible even if the statistical noise of the measured hologram in each individual pixels exceeds the holographic signal.

As a result, the variance of the amplitude of the image $(U(\mathbf{R}))$ caused by statistical noise is given by

$$\sigma^2_{|U(\mathbf{R})|} \approx \frac{I_{tot}}{N^2}$$

Here N is the number of measured data points forming the hologram and $I_{tot}$ is the total number of counts collected. Then the signal-to-noise ratio (SNR) of the reconstructed image at the position of an atom can be expressed as

$$SNR \approx \sqrt{2I_{tot}}\,\frac{b}{R}\,X(\mathbf{R})$$

where $X(\mathbf{R})$ is a factor taking in to account the twin effect and ranging from 0 to 2.

Thus reconstruction of the measured hologram can be successfully carried out even at a noise level in $\mathbf{k}$-space which largely exceeds the holographic oscillations, i.e. when $SNR \geq \sqrt{1/(2N)}$ ($N$ denotes the number of measurement points) provided the Nyquist criterion for the resolution is satisfied.

## Some experimental results

The internal source concept was first proven by a Canadian group [4] using for sample a mineral crystal called Simpsonite. Later on successful holographic experiments were carried out on PdH alloy [5] and ammonium chloride [6].

The internal detector approach was realized on $Pb_{99.74}Cd_{0.26}$ alloy [7]. An unexpected deviation between the calculated and observed intensities of the reconstructed holographic images was observed (see Figure 3.)

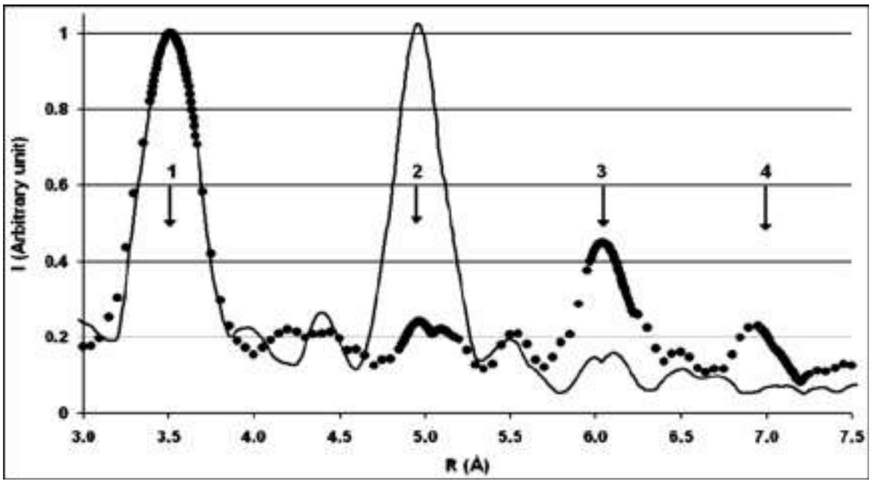


*Figure 3. The intensity distribution of the reconstructed holographic image as a function of the distance R from the Cd nucleus. Experimental values (●) and model calculation for a non-distorted crystal involving 100 surrounding shells (continuous line).*

This deviation can be interpreted by supposing that in the vicinity of the Cd atom the lead lattice gets distorted. Using the cosines rule the degree of the distortion was calculated with extremely high (picometry) precision.

## The double reconstruction

As it was discussed at the beginning part of the present talk hydrogen fulfil the role of point-like source of neutrons. However, it is easy to see that the number of neutrons at the position of hydrogen nuclei - protons - is modulated by carrying out the internal source experiment.
These neutrons are scattered to the detector synchronously with those which form the internal source hologram. In some sense the hydrogen nuclei play the role of internal detector. These secondary neutrons appear as contamination in the primary internal source hologram causing additional oscillation in it. At the first sight there is no way to get rid of such secondary scattering contribution. Nevertheless by applying an ingenious solution this drawback can be turned even into the advantage. Let consider the following arrangement as it is shown in the Figure 4.

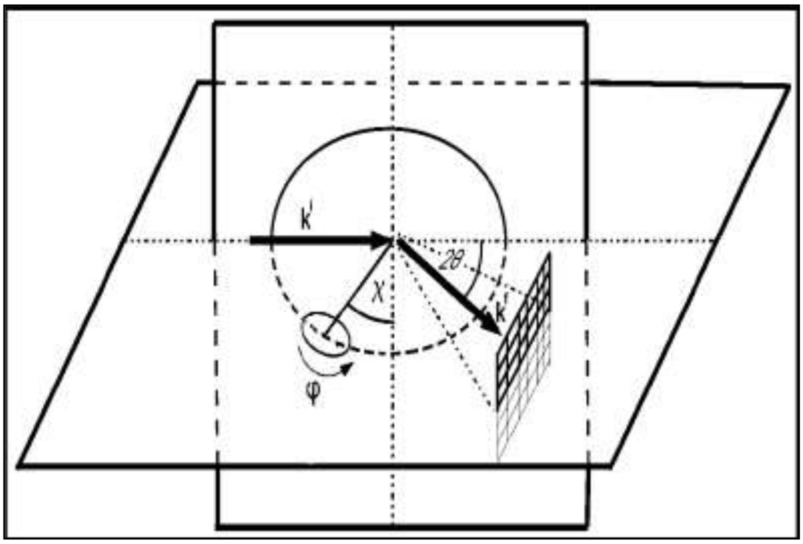


*Figure 4. Schematic of a measurement set-up using a 2D PSD. The angle2Θ is fixed (the detector is not moving)*

This set-up was applied in an experiment on $PdH_{0.51}$ system [8]. The two-dimensional (2D) area detector was installed by such a way that it covered the angular region between the direct beam and the first Bragg peak. By this way we avoid the high redundant count rate. The collected data were treated by two different ways. At each pairs of χ,φ angular position the total collected events are summed up, i.e. the data collection is considered as it were an internal detector experiment.

It is shown that the observed intensity can be written as:

$$I\left(\bar{k}^i, \bar{k}^f\right) \approx \frac{I_0^i}{R^2}\left(1 + \chi^i\left(\bar{k}^i\right) + \chi^f\left(\bar{k}^f\right)\right)$$

In other words, the observed holographic intensity is represented as the sum of IDH and ISH. It is important to note that the vectors $\mathbf{k}^i$ and $\mathbf{k}^f$ are defined in different vector spaces. The first one is the wave vector of the incoming beam, whereas the second one is the wave vector of the measured beam relative to the sample orientation. Thus, changing one of the rotation angles of the spectrometer entails different changes of the two vectors. The space of $\mathbf{k}^p$ corresponds to the IDH space and the space of $\mathbf{k}^s$ to the ISH space.

The reconstruction of the data set is then treated by Barton's procedure in the momentum space of the incoming neutrons. In the second treatment the counts collected in each separate pixel is considered and the reconstruction is carried out as in the internal source case. Both above described procedure give rise to identical but independent holographic image. Summing up of the results of the two separately obtained hologram one gets a doubled value of the holographic signal at the same statistical noise level. So, the signal-to-noise ration is increased by a factor of two. Moreover, the perturbation of the holographic image caused by secondary neutrons is converted into a useful signal and thus they anymore do not act as a contamination. By other words in the resulting hologram the oscillation caused by contamination is in large extent suppressed and the picture gets much cleaner (see Figure 5). In addition, since the position of the detector favours the forward scattering the perturbations stemming of the diffuse scattering is significantly suppressed.

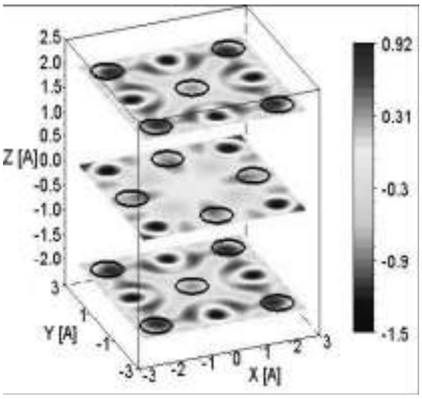


*Figure 5. The amplitudes of the holographic image of $PdH_{0.51}$ reconstructed by the DR method. The layers of the reconstructed images are parallel to the (001) lattice planes at z = −2, 0 and 2 Å, respectively. The positions of the 6 first and 8 second neighbour Pd atoms (marked by black circles) and the 12 first neighbour H atoms (marked by white circles) arranged around the hydrogen atom at the centre of the coordinate system (not shown) are indicated. The spots surrounded by black circles represent the Pd atom positions and the spots give the positions of the H atoms. The centres of the circles coincide with the atomic positions of an ideal, undistorted crystal lattice. Two weak spots in the middle of the upper and lower planes in (d) hint at the presence of first neighbour Pd atoms.*

## Conclusions and future prospects

The author hopes to have been able to convince the reader that atomic resolution neutron holography (ARNH) is a promising new tool for studying the 3D local structure of crystalline materials with extremely high accuracy. Existing spallation neutron sources (ISIS, SNS, JSNS) are powerful pulsed sources of neutrons. It is expected that in the not too distant future the European Spallation Source ESS will also come into operation. As it is well known, so-called time-of-flight techniques allow making use of the total quasi-Maxwellian spectrum of neutrons produced by the target of pulsed sources. By exploiting this opportunity, multi-wavelength ARNH using the double reconstruction method (DR) together with position sensitive area detectors will become feasible. In addition, preliminary model calculations show that ARNH can be extended for study of polycrystalline samples provided if the investigation is satisfied only with the knowledge of the distance between the source/detector atom and the neighbouring ones.

Finally, it should be noted that the high neutron beam intensity of the mentioned new sources will allow extending the application of ARNH to samples consisting of nuclei for which the cross sections both for incoherent scattering and absorption are rather small. This offers a wide range of applications in materials science – including the investigation of magnetic systems – in the not too distant future.

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