Atomic resolution neutron holography (present status and future prospects)

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Abstract

The idea of atomic resolution neutron holography proposed by the authors of the present talk has been experimentally proven using both the internal source and internal detector concept in three various experiments carried out on samples simposite, PbCd alloy and PdH metalhydride. It was shown that the 3D image of the local atomic arrangement around the source/detector the position of nuclei could be with high accuracy (better than 0.01 Å) reconstructed. This accuracy allows observing the local lattice distortion of the lattice by a direct way. In order to achieve these results numerous corrections (e.g. getting rid of Bragg reflections, taking into account of the effect of twin images, the influence of the far removed objects on the holography images and the distortion caused by the finite resolution of the instrument used) should be properly applied. In the present talk we discuss the way for extending of systems to be investigated. Some methodical news (e.g. the use of multiwave holography, the investigation of not fully ordered systems) are also considered.

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1. Introduction

In his pioneering work Abraham Szőke \cite{1} established the basic concepts of the atomic resolution holography using electromagnetic radiation and slow electrons. These ideas had been soon experimentally proven \cite{2,3}. In principle, any de Broglie waves can be applied for that purpose. There are two ways of carrying out the observation of atomic resolution holography. Either the emitter of the radiation used is embedded in the sample, this is the so-called internal source concept, or relaying on the optical reciprocity law the detector is a part of the sample. This last approach is called as internal detector concept.

2. Why neutrons?

Comparing neutron holography with electron and X-ray holography one can discover several features limiting the application of each all of three types of irradiation. The main difference takes its origin from the fact that electrons and X-rays interact with the electrons of the sample while neutron interacts with nuclei. Moreover, electrons interact with the condensed matter strongly, thus, they are restricted essentially to the investigation of surfaces. X-ray being able to penetrate more deeply into matter interacts with the electrons but their use for many systems is unfavourable either due to the too small or too high value of the electron density. In the first case the scattered signal is too weak while in the second case the self-absorption of the sample gives rise to distortion of the scattering pattern requiring serious corrections. The atomic form-factor, i.e. the electron density distribution in the atoms makes the electron and X-ray pattern more complicated for interpretation. Thermal neutrons, in
principle, are free of these drawbacks. Moreover, they possess many well known attractive properties. However, the limited intensity of the neutron sources and the relatively small scattering amplitudes may create obstacle to realization of atomic resolution neutron holography. These obstacles make the task of discovering of the way of the use of thermal neutrons even more challenging. A particular difficulty arises because no nuclear reactions are available in the course of which thermal neutrons are emitted from a point like source. This difficulty can be evaded using nuclei embedded in the sample, which can redistribute the illuminating neutron beam into 4 π solid angle with high efficiency. This condition is satisfied by the proton [4]. That is the internal source concept can be performed in this way. The criteria of the internal detector concept can be easily satisfied using nuclei possessing high neutron capturing cross-section and the capturing event is promptly followed by emission of gamma- or beta-rays. Very soon after appearing of our first paper [4] these ideas got impressively proven in several experiments [5–7]. The internal source concept was demonstrated in an experiment on a mineral (a single crystal of natural simpsonite with the chemical composition \( \text{Al}_4\text{Ta}_3\text{O}_{13}(\text{OH}) \)) by a Canadian group [5] and in an another experiment performed by the present authors [7] on the metal-hydrogen system \( \text{PdH}_{0.78} \).

The internal detector concept was tested on the \( \text{Pb}_{0.9974} \text{Cd}_{0.0026} \) crystal. The yield of gamma-rays emitted by Cd nuclei following neutron capture was registered as the function of the sample orientation [6]. These achievements are illustrated in Figs. 1 and 2. It is worth to note that while the separation of O atoms in Canadian experiment [5] was found with accuracy of 0.3 Å from the mean, the Pd–H distance in [7] was found with accuracy of 0.1 Å. The Pd–Cd separation [6] was obtained with accuracy better than 0.01 Å. For achieving such an accuracy the distortion effect of the far removed nuclei (up to 102 shells) was taken into account.

3. Perspectives

3.1. New results expected

Till now atomic resolution neutron holography is considered as a phenomenon. The next step ahead is converting this phenomenon into an instrument to be applied in various field of solid state physics and material sciences. The above results hold some promise for future practical applications. First, there is a large number of substances containing substantial amounts of hydrogen such as metal-hydrogen systems, numerous organic substances and many minerals of geological importance. Secondly, the recording of holograms based on magnetic scattering may open up a new perspective on the investigation of magnetic materials. Holographic experiments are providing much structural information. The neutron hologram recorded in both the inside-detector and inside source experiments provides unambiguous evidence about the local environment of a given atom in a crystal. This will be used for following up the phase transitions, or look after what interstitial site is occupied by a hydrogen atom e.g. in metal hydrates. The area of application of the internal source approach is extremely wide. Amongst others, the knowledge of the position of hydrogen atom is important not only for the study of simple systems as metal hydrates. Determination of the position of crystalline water molecules in crystals especially of biological origin has a particular interest. In systems containing too much hydrogen at various sites, some of protons can be replaced for deuterons. Such a replacement left the molecules from chemical point of view unchanged while for the neutron holographic experiment provides excellent contrast. The study of the hydrogen bonds playing basic role in the spatial formation of structures of many complex molecules also can be included in the repertory of the neutron holography.
Many isotopes available exhibiting reasonably high absorption cross sections are warranting their use in inside detector experiments. Almost all of the elements with \( Z \geq 45 \) have isotopes possessing absorption cross sections close to 5 barn or higher. For \( Z \leq 45 \) about 15 isotopes fulfill this condition. This situation promises that a wide variety of compounds and alloys are potential targets for neutron holographic investigations using the inside detector approach. Owing to the small size of the scattering centres (i.e. the size of atomic nuclei) the interatomic distances can be measured with extremely high accuracy (better than 0.01 Å). Thus, the direct measurement of the local distortion caused by an impurity atom becomes possible. This information serves for much deeper understanding of the lattice deformation.

The crystal lattice can be restored from the hologram without making use of any a priori knowledge about the orientation of the sample. For example, using neutron holography the reconstruction of the position and orientation of pre-determined groups of atoms in a large and complex system can be carried out [8]. The method based on pattern recognition permits to measure the spatial and angular coordinates of the group. Fourier hologram of the unknown compound object is taken for the initial signal and the hologram of a group under investigation is used for the reference one. The magnitude of the cross-correlation function of the pattern hologram to the measured one possesses a series of well-determined maximum that characterize the probability to find the pattern in the molecule studied. This approach has particular interest for complex bio-molecules like proteins composed of stable groups of atoms (amino acids) which spatial configuration and position in the molecule define the properties of the molecule.

3.2. Methodical improvements

In order to achieve the above enumerated goals one needs profound improvements of the methodical background of the holographic experiment. One of the major technical drawbacks for wider applications of neutron holography is the rather long measuring time (several days) required to record a hologram. The use of multidetectors instead of single one improves the situation significantly. Gains of several orders of magnitude over a single detector set-up can be envisaged in the near future. The new high flux spallation neutron sources (US-SNS, J-SNS and the ESS) under construction promise double benefit. Besides the increase of the intensity of the source these pulsed sources allow to carry out holographic measurements with multi-wavelength neutron beam simultaneously. For centrosymmetric samples the real and twin images of the atomic nuclei appear at the same positions. This superimposition at a given wavelength causes partial or even full cancellation of the holographic spot imaging the object. However, at another wavelength the condition for cancellation is no longer satisfied. A multi-wavelength experimental set-up automatically provides the condition for finding any cancelled spot. The time-of-flight technique combined with the multidetector data collection would reduce the measuring time of the neutron holography experiment down to several hours, or even minutes.

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