

Atomic resolution neutron holography
PhD Thesis

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Overview of the research field

The holography invented by Dennis Gabor became capable for atomic resolution imaging by the idea of Larry Bartel and Ábrahám Szöke. Their method is based on the use of single atoms as secondary electron source. In the reconstructed hologram the local neighbourhood of the source atom is seen. This method is called as inside source holography. The method was applied for X-ray radiation by Gyula Faigel and Miklós Tegze. Later on, Gog, applying the rule of optical reciprocity, carried out similar experiments, using macroscopic source and atomic detector instead of atomic source and macroscopic detector. This method is called as inside detector holography. László Cser proposed to use both method applying neutron radiation. In his idea the atomic neutron source is a strong incoherent scattering centre. The optimal choice is the proton which has the strongest incoherent scattering among the known isotopes. He proposed also strong neutron absorber isotopes emitting prompt but not late gamma photons as detector isotopes. This type of holography is named as gamma conversion neutron holography (GNH) The first inside source neutron holographic experiment was carried out by Sur and his co-workers, while the first inside detector neutron hologram was measured by László Cser and his co-workers. The proton can act not only as the source of the neutrons scattered incoherently on it, but also as the detector of the neutrons of the direct beam, and the primary beam scattered coherently by the sample. Thus, depending on the measurement geometry, the inside source hologram, inside detector hologram or both can appear in the measured image (this problem is known in X-ray holography also). I call these measurements collectively as incoherent neutron holography (INH).

The atomic resolution hologram has serious advantages comparing to diffraction. The centre of the hologram is fixed (it is the position of the source/detector atom), and the amplitude of the reconstructed holographic image is inversely proportional to the distance

from the origin. Thus the hologram shows the local neighbourhood of the source/detector atom, i.e. it shows directly the partial pair-correlation function. Diffraction image, however, is defined by the total pair-correlation function. Moreover, the phase problem of the diffraction is missing in holography. Thus the position of an atom in the unit cell, or the local distortion of the host lattice due to the presence of an impurity atom can be directly measured by using holography. In spite of these advantages of atomic resolution holography, it did not become routine measurement method. Except some measurement, the aim of the experiments were to show the possibility of different holographic measurement methods, but not to answer serious questions of materials science or solid state physics. The most important informations for carrying out successful holographic experiments like the necessary resolution, calculation method of signal-to-noise ratio of the reconstructed image, description of artifacts, and the methods to avoid them are missing or just partially discussed in the literature.

The aims of the work

The primary goal of my PhD work is the correct mathematical description of the atomic resolution neutron holography. One part of it is the determination of the criteria of the successful holographic measurement. For this work, the effects of the non ideal measurement (finite measurement range, finite sampling, finite measurement time) in the reconstructed image is to be calculated. The final goal of the theoretical work is to calculate optimization functions, elaborate measurement procedures and new methods, and give methods for retrieving the total structural information from the hologram.

Furthermore, my goal is also to refurbish the thermal three axis instrument of the Budapest Research Reactor being capable to make optimal holographic measurements considering the results of the the-

oretical work.

Finally, the practical aim of the work is to carry out holographic measurements on ammonium-chloride (NH_4Cl), palladium-hydride and tin-cadmium single crystal samples so in Hungary as in foreign neutron centres to validate the calculations, and to answer real physical questions.

New scientific results

I collect the results of my PhD work in the following theses:

1

I calculated the effects of the resolution of the instrument, the mosaicity of the sample, and the thermal movement of the atoms on the hologram. I proved that the reconstructed peaks of the atoms are shifted by not only the statistical noise but also by the side oscillations of the images of the neighbouring atoms. I improved the holographic model by calculating the effect of the finite resolution. The new model became absolute convergent, which enables more accurate calculations by increasing the number of atoms in the model. I gave an approximative function to calculate the minimal distance from the origin of the reconstructed hologram where false peaks appear due to the finite sampling (Aliasing effect), and to calculate the amplitudes of these false peaks. [T1, T2]

2

I calculated the signal-to-noise ratio of the reconstructed hologram, and the effects of the different image improving methods on it. I proved that the signal-to-noise ratio of the reconstructed image is proportional to the total count of the measurement but not to the

counts of one measurement point. I proved that the symmetry operations are increasing the signal-to-noise ratio of the reconstructed image. Using these results, I gave an optimization method for increasing the signal-to-noise ratio. The method can be applied in every type of atomic resolution holography independently on the applied beam. [T3]

3

I gave a new method (named double reconstruction method, DR) for INH, where the inside source and inside detector signal defines collectively the measured image. In this method one measure the two signal in one measurement, and reconstructs them separately. The DR method causes significant decreasing of the artifacts of non holographic effects (like elastic diffuse scattering), and it improves the signal-to-noise ratio. Moreover, the DR method gives intrinsic solution to the determination of the position dependent efficiency of the two dimensional position sensitive detector without additional efficiency measurements. The results are proved experimentally (see theses 5th and 7th). [T4]

4

I built a dedicated holographic instrument at the 8th thermal neutron channel of the Budapest Research Reactor by refurbish a three axes instrument. The instrument is capable to measure so gamma-conversion internal source hologram as incoherent neutron hologram. The resolution of the instrument can be set to optimal if the needed maximal range of visibility is not smaller than 2\AA . This is the first (and until now unique) dedicated neutron holographic instrument in the world. [T5]

5

I carried out successful incoherent neutron holographic measurement and reconstruction on ammonium-chloride (NH_4Cl) single crystal. The experiment proved the efficiency of the holographic instrument (4th these) and also the efficiency of the double reconstruction method. The result of the measurement shows that the local neighbourhood of the H atom is in good agreement with the Frenkel model. [T5]

6

I carried out gamma conversion inside detector holographic experiment and successful reconstruction on tin-cadmium single crystal. I proved that the cadmium atoms are in substitutional place in the tetragonal host lattice of the tin. The experiment proved also that the holographic instrument (4th these) is capable for GNH measurements. [T5]

7

I measured successful INH experiment on palladium-hydride system. The experiment proved the advantages of the DR method with two dimensional position sensitive detector (3th these). Moreover, the measurement proved that it is possible to make neutron holographic measurement even with high temperature sample environment. [T4]

8

As a result of the investigation of the holograms of the systems possessing central holographic symmetry I gave a high accuracy data treatment method for calculation the distance of the neighbouring atoms. I applied this method on the first GNH measurement (on lead-cadmium alloy). I determined the local distortion in the lead lattice caused by the cadmium atom with picometer accuracy. The

result showed that the first neighbours of the cadmium are farther than in the undistorted lead lattice in spite that the atomic radius of the cadmium is smaller than that of the lead atom. However the mean of the distortion of the first four neighbours shows decreasing. The shift of the position of the first four neighbours are in qualitative agreement with the oscillation of the charge density caused by the difference of the numbers of valence electrons of lead and cadmium (i.e. Friedel oscillation). [T2, T6, T7]

Publications referred in my thesis points

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- [N4] A. Szakál, Sz. Czifrus, M. Markó, J. Füzi, L. Rosta, and L. Cser. Optimization of focusing supermirror neutron guides for low [gamma]-background. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 634(1, Supplement 1):S130 – S133, 2011. Proceedings of the International Workshop on Neutron Optics NOP2010.