Experimental observation of neutron holography on atomic level

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Abstract

As an alternative to X-rays and electrons a recent paper proposed the use of thermal neutrons with wavelengths close to interatomic distances in condensed matter to obtain holographic images on an atomic scale. Two experimental methods were considered which either put the radiation source inside and the detector outside the object or vice versa. The second approach, known as the inside-detector concept, requires strongly neutron-absorbing isotopes acting as point-like detectors in the sample. We demonstrate the feasibility of this technique for the first time by recording a holographic image of lead nuclei in a Pb(Cd) single crystal

Introduction

Holographic techniques, well known from the case of visible light,[1] have been successfully extended to X-rays and electrons during the last decade resulting in images providing atomic resolution[2,3]. Alternatively, the use of thermal neutrons with wavelengths close to interatomic distances in condensed matter has recently proposed[4] [L.Cser, G.Krexner, and Gy.Török, *Europhys. Lett.* 54, 747 (2001) to obtain holographic images on an atomic scale.

In the first of two approaches put forward, one is known as *the inside-source concept,* nuclei like hydrogen exhibiting large incoherent neutron scattering cross section serve as point-like sources of neutron spherical waves.

The second approach involves the inside-detector concept inter-changing the positions of source and detector as suggested by the principle of optical reciprocity. The name of this technique derives from the fact that the neutron *source* is located *outside* the object to be imaged while the *detector* is placed *inside* the sample

1.) The point-like inside source concept

This method requires a point like inner source of monochromatic neutrons. Such a source can be created using a point-like scatterer of the neutrons possessing extremely high incoherent scattering cross section. [4]. Incoherent scattering redistributes the incident neutron beam into 4π solid angle isotropically. It is well known that the incoherent proton – neutron scattering cross section σ_{inc} is equal to 79.91 barns, which value may by about two orders exceed σ_{inc} of many other nuclei.

Elastic multiple scattering on the protons does not cause any problem since in the course of this process the scattered neutron always meets a successive „redistributing" proton and thus can be used again.

The point-like inside source principle is illustrated in the Figure 1.

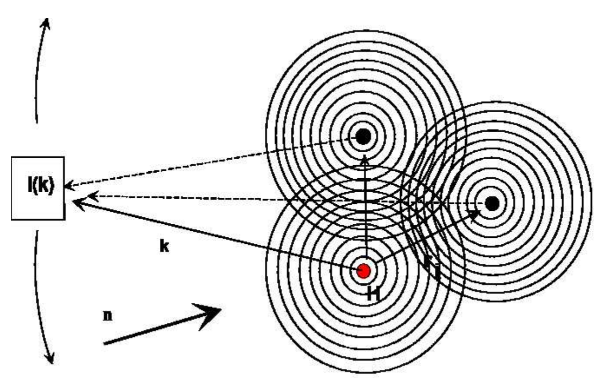

Figure 1. The monochromatic incident neutron beam illuminates the sample. Neutrons incoherently scattered by hydrogen nuclei form spherical waves. These waves partly are scattered by neighbouring nuclei and partly reach the detector located at large distance. The detector moving over a sphere around the sample records the interference pattern of these waves.

2.) The point-like inside detection concept

Neutron holography can be done by an alternative way using the principle of the optical reciprocity [5,6]. In this method the positions of the source and the detector are interchanged. The plane wave from a far-field source reaching directly a particular nucleus in the sample can be considered as the reference beam, while the waves being scattered from neighbouring nuclei serve as object beam.

The experimental layout in this case will be extremely simple (see Figure 2). The samples mounted on a proper goniometer will be put into a well-collimated and duly monochromatized neutron beam. The gamma rays following the neutron capture will be detected by the use of a scintillation detector installed in such a way that the part of the sample illuminated by neutrons is visible (Bragg geometry). This set-up is advisable because bulk samples would considerably attenuate the intensity of the gamma - rays. In most of case energetic characteristic X-rays will be generated by the gamma rays. Since each element is largely transparent for its own characteristic X-rays the capturing event can be efficiently registered even via the detection of this radiation. Nevertheless, the photo-effect will attenuate this radiation yet.

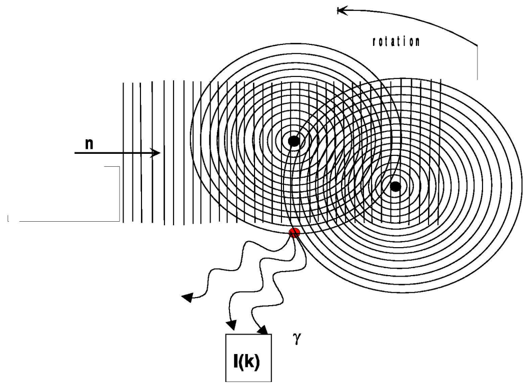

Figure 2. Incident monochromatic neutron wave emitted by a distant point-like source propagates towards the target nucleus forming the reference beam. The scattered part of the wave serving as object beam interferes with the reference one. The resulting neutron density modulation is converted into prompt –gamma rays by the nucleus. These gamma-rays are registered by the detector I(k) .

For experimental realization of this later approval a well collimated neutron beam, i.e. a plane wave, propagates towards a single-crystalline sample containing a small amount of atoms whose nuclei are strongly neutron-absorbing. The incoming neutron wave reaches these detector nuclei either directly, without any scattering (and may thus serve as the reference beam), or after scattering from other nuclei in the sample. This latter process gives rise to the object beam.

In the work of *L. Cser, Gy. Török, G. Krexner, I. Sharkov, and B. Faragó (Phys.Rev.Lett.89, 175504 (2002))* [8] the feasibility of the inside-detector technique is demonstrated for the first time by recording a holographic image of lead nuclei in a Pb(Cd) single crystal. The obtained result is on Fig.3. displayed

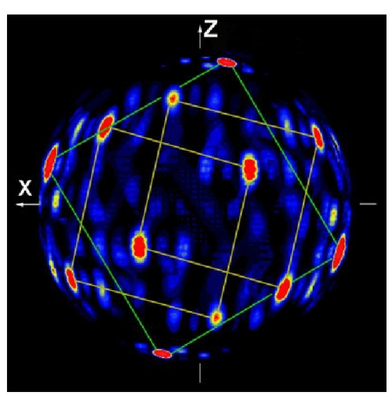

Figure 3A. The spots representing the positions of the twelve Pb atoms forming the first neighbours of the Cd nucleus.

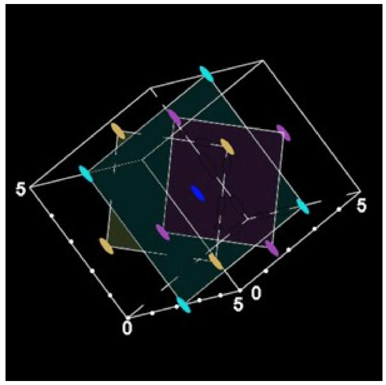

Figure 3B. Schematic illustration of the arrangement of the lead atoms surrounding a Cd atom (yellow spot) as first neighbours in the FCC structure. The orientation and positions of the spots are equivalent in Figure 3A.

The inside-source concept has already been successfully applied recently *[7] B.Sur, R.B.Rogge, R.P.Hammond, V.N.P.Anghel, and J.Katsaras, Nature 414, 525 (2001)*

Further the holographic image of the atomic arrangement in a $PdH_{0.78}$ single crystal also was recorded [9] using spherical neutron waves generated by incoherent neutron-proton scattering ('internal source concept'). The resolution is sufficient to show the positions of single atomic nuclei on their respective lattice sites despite the delocalization of protons due to the weak bonding of hydrogen dissolved in palladium metal. Ways to overcome problems in the reconstruction of atomic positions related to the highly unfavorable sample shape are discussed. The technique offers new possibilities for the investigation of various materials containing substantial amounts of hydrogen The result of reconstructed hologram is on the Fig 4. displayed

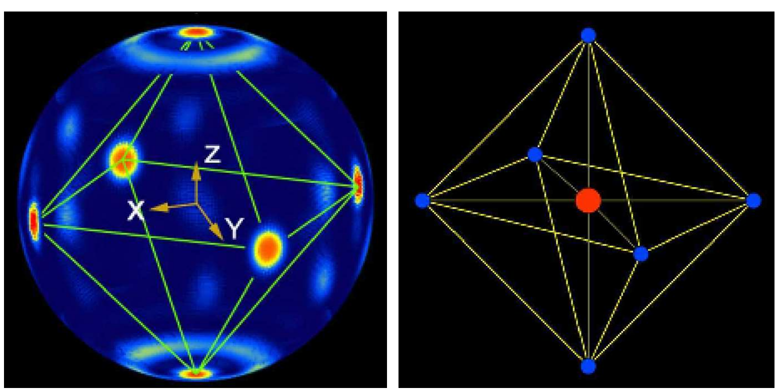

Figure 4. The restored hologram using represented in spherical coordinates. The z-axis is parallel to the (100) crystallographic direction.

The hydrogen atoms in the $PdH_{0.78}$ system occupies the octahedral interstitial positions in the Pd crystal. For the orientation chosen the model calculation predicted that the presence of six first neighbours in the reconstructed image will be represented by four spots in the equator and two spots at the upper and lower poles correspondingly. The observation of six spots in the reconstructed hologram corresponds to the expectation that hydrogen is sitting in the octahedral interstitial position in the palladium crystal. This conclusion is supported by the fact that the positions of the spots correspond to the orientation of the sample.

From the calculation of the centres of the spots observed the atomic distances between the hydrogen and palladium atoms were estimated. The obtained value is 2.04 ± 0.1 angström in good agreement with the crystallographic data.

Contrary to the case of X-rays the size of the reemitting neutron source is small. The apparent size is given by the zero-point oscillation, i.e. by the finite value of the localization of the proton being on the ground level of the potential well. Elementary estimation predicts the localization of the proton in PdH to be within 0,3 angström. The size of the scattering objects, i.e. the size of the neighbouring nuclei is even smaller (a few *fm*). So, it is expected that the size of the spots observed will be formed by two contributions. First, by the resolution power of the instrument and second, by the size of the source.

From our observation it follows that despite of the low energy distance between the excitation spectrum of the hydrogen in palladium the considerable part of the neutrons are scattered elastically.

Conclusions and future prospects

Now, relaying on the two independent experiments presented above the feasibility of both concepts of atomic resolution holography proposed in Ref.4 is proven.

Holographic experiments are providing many structural information. From our inside detector experiment it follows that neutron hologram gives evidence that Cd atoms are occupying the Pb sites in the alloy. The atomic distances between the detector Cd nucleus and the neibouring Pb ones were determined with accuracy of few hundreds of angströms.

Due to the small size of the scattering centres (i.e. the size of atomic nuclei) the interatomic distance can be measured with extremely high accuracy. This accuracy is limited only by collimation and monochromatization conditions of the neutron beam used.

The hologram can be restored without use of any *a priori* knowledge about the orientation of the sample.

On the basis of these results practical applications of atomic-resolution neutron holography appear to be highly promising. The special properties of the nuclear scattering process of neutrons, including its isotopic sensitivity and its dependence on the magnetic moment, will enlarge the field of investigation opened up by the use of neutron holography of which we briefly mention just two examples:

First, the internal-source concept is applicable to the investigation of a wide variety of hydrogen-containing compounds, both anorganic and organic.

Second, by using polarized neutron beams both the internal-source and the internal-detector concept may be extremely useful and flexible tools for studying the local magnetic structure of magnetic materials.

Third, thanks to the very small size of the atomic nuclei neutron holography provides an unparalleled accurate and direct measurement of the local crystal lattice and magnetic structure distorsion.

Some advantages of applications of neutron holography:

Take advantage of the complementarity of neutrons to tackle those systems that cannot be easily studied with electromagnetic radiation. Neutrons are coherently scattered equally well by light and heavy atoms Neutrons can distinguish similar-Z atoms and can distinguish some isotopes.

The internal source H atom is abundant in biological materials and polymers. The Hydrogen and Deuteron scatters very differently. Neutrons are gentle, causing little or no damage to delicate (biological systems due to a small incoming neutron energy (a few meV))

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