Importance of Neutron Scattering for Materials Science: Expectations on Magnetic Holography

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The neutron scattering technique is an indispensable probe for structural investigations of materials, particularly, for observations of light atoms and magnetic moments. Using the IMR neutron powder diffractometer HERMES, which can observe scattering patterns in wide Q range at once, we have succeeded in performing neutron holography experiments in a Pd-H compound. Holography will become a quite important technique for local structure investigations of materials. Moreover, as the next stage, magnetic holography with polarized neutrons must be promising. [DOI: 10.1380/ejssnt.2011.422]

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I. INTRODUCTION

The neutron scattering technique has many important characters for materials science: high sensitivity of light atoms and spins, capability of accurate observations of spin-lattice dynamics, larger penetration depth and so on. Since neutrons are effective for almost all the elements, the neutron scattering technique is indispensable to create breakthroughs in novel material science. For material science, structural investigations of atoms and spins are important to understand the origins of properties of materials. Neutron diffraction experiments are the best way for such structural investigations; in fact, it is widely used for magnetic structure refinements, determinations of positions of light atoms, visualisation of pass of ion conductors, and so on. However, the diffraction technique can give us information on only averages of periodic structures of atoms and spins, and cannot obtain information on local structures around specific nuclei. Moreover, since the diffraction technique intrinsically includes so-called “the phase problem”, structural refinements always need highly reliable structure models. From this point of view, the neutron holography technique will be an important technique which provides information on detailed local structures of light atoms, such as H, O, N, Li, and spins, without any structure models.

This article gives general explanations about importance of the neutron scattering technique in material science. In Section II, general characters of neutrons for material science will be given. In Section III, short explanations of neutron facilities and instruments in Japan will be given. Typical data of the neutron powder diffraction technique will also be shown. Moreover, in section IV, we will explain importance and expectations of neutron holography, which is developed on Kinken (Institute for Materials Research) neutron diffractometer. Detailed of the holography experiments on HERMES is reported elsewhere in this symposium by Hayashi et al.

II. GENERAL CHARACTERS OF NEUTRONS

The neutron diffraction technique is one of the most important probes for structural investigations in material science, as well as the X-ray diffraction technique. Some characteristic points and advantages from points of view of material science are shown below.

1. Neutrons have large penetration depth to materials because of electric neutrality; thus, neutrons can observe bulk properties of materials. Moreover, non-destructive observations of industrial products, such as engines of automobiles, wings of airplanes, are possible.

2. Scattering length is independent of atomic number because neutrons are scattered by atomic nucleus, not by electrons. Thus, the neutron scattering technique has advantages in distinction of atoms with closed atomic numbers, and accurate observations of light atoms, such as H, C, N, O, in materials which include heavy atoms.

3. Since a neutron has spin (S = 1/2), magnetic response in magnets can be observed by the neutron scattering technique. In particular, neutrons have high sensitivity on determinations of magnetic moment alignments. Note that this makes neutron magnetic holography possible.

4. The wave length and energy of thermal and cold neutrons, which are used in standard scattering experiments, are quite close to the scales of spatial and time correlations of atoms and spins in materials, meaning that information on spatial and time correlations can be obtained accurately and simultaneously by the neutron scattering technique. Thus, the neutron spectroscopy is indispensable to understand interactions in materials qualitatively.

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5. Sample preparation is relatively easier than those for X-ray experiments because preferred orientation effect, particle size effect, surface effect, are not serious for neutron experiments.

6. Experiments under extreme condition such as low temperatures (~10 mK), high static magnetic filed (~13 T), are relatively easier. Based on these advantages, the neutron scattering technique is a useful probe of novel material science, in particular, environment functional materials, such as Li batteries, fast ion conductors, hydrogen storage materials, and magnetic materials such as high-Tc superconductors, multiferroics materials and so on.

On the other hand, it should be pointed out that neutrons have some weak points, such as:

- For neutron scattering experiments, large amount of samples (a few cc or more) and long measurement durations are needed in comparison with X-ray experiments. This is particularly serious for neutron holography because a lot of measurement points are required to obtain a hologram image.

- For materials which include some elements, such as Cd, B, Gd, Sm, Eu, neutron scattering experiments are impossible or extremely difficult because of the strong absorption effect. For experiments of such materials, the replacement to isotopes is needed. Unfortunately, most of isotopes are expensive.

- Though the neutron scattering is an accurate probe for observation of hydrogen, crystal structure refinement of hydrides by neutron powder diffraction are difficult in some cases because of high incoherent scattering from hydrogen itself, which makes effective S/N ratio quite worse. For improvement of the S/N ratio of hydrogen samples, deuterization is a good way, though the isotope exchange effect in hydrides may not be negligible.

To overcome these disadvantages, therefore, complementary investigations with radiation X-ray, μ-SR and other probes must be quite effective.

III. FACILITIES AND DIFFRACTOMETERS/SPECTROMETERS

There are two kinds of neutron facilities: accelerator facilities and reactor facilities. A main reactor facility for material science in Japan is JRR-3 of Japan Atomic Energy Agency, and the accelerator facility in Japan is the Materials and Life Science Experimental Facility (MLF) in Japan Proton Accelerator Research Complex (J-PARC). Roughly speaking, accelerator facilities are good at wide-Q range measurements with higher resolution, and high energy spectroscopy (> 80 meV), while reactor facilities have an advantage for detailed observations at a particular point in the Q-ω space. Moreover, since J-PARC will be the most brightest neutron facility in the world, difficult experiments for which very high neutron flux is required, such as holography experiments, will be feasible in J-PARC.

From a point of view of the holography technique, reactor facilities must be suitable because monochromatic beams are usually used in reactor facilities, while white beams are used in accelerator facilities; note that monochromatisation of neutron is an essential point for the present neutron holography technique. Figure 1 is Kinken powder diffractometer HERMES [1], which is a typical powder diffractometer in reactor facilities; HERMES is used for the neutron holography experiment project in IMR. HERMES has a banana type detector bank in which 150 neutron detectors with an angular interval of 1° are installed. Using the multi detector system, user can observe diffraction patterns in wide Q-range (0.1-7 Å⁻¹). This makes it feasible to obtain a neutron hologram image during relatively short duration.

FIG. 1: Kinken (IMR) neutron powder diffractometer, HERMES, installed at JRR-3 of Japan Atomic Energy Agency, Tokai, Japan. HERMES has a banana-type detector bank in which 150 neutron detectors with an angular interval of 1° are installed. Using the multi detector system, user can observe diffraction patterns in wide Q-range (0.1-7 Å⁻¹). This makes it feasible to obtain a neutron hologram image during relatively short duration.
IV. NEUTRON HOLOGRAPHY PROJECT IN IMR

In IMR, neutron holography experiments are in progress on HERMES. There are some precedent experiments of neutron holography using incoherently scattered neutrons by Sur et al. [2] and Cser et al. [3]. On HERMES, we have succeeded in obtaining hologram images of a Pd-H single crystal as well [4].

Here, the authors give a brief explanation of the principle. Details are shown in Ref. [4]. Incoherently scattered neutrons from atoms are important to create coherent waves in holography. This is based on the inside-source concept in X-ray holography, where nucleus, such as hydrogen, with large incoherent neutron scattering cross sections play a role of point-like sources of neutron spherical waves, analogous to fluorescing atoms in X-ray fluorescence holography. The intensity of the wave with a \( k \)-vector and at a distance \( R \) is given by

\[
I_h(k) = \frac{I_0}{R^2} \left\{ 1 + 2Re \left[ \sum_i \frac{b_i}{r_i} \exp\{i(r_i k - r_i k)\} \right] \right\},
\]

(1)

where \( I_0 \) is the incident intensity, \( b_i \) and \( r_i \) are the scattering length and the position vector of the \( i \)-th nuclei with respect to the source, respectively [4]. The second term in Eq. (1) corresponds to holographic oscillation. Since the ratio of the signal to background intensities in the hologram is on the order of \( 10^{-3} \), collection at one million counts in each pixel is adequate. If one uses a single counter diffractometer, measurements time for holography will be unfeasibly long. HERMES of IMR, on the other hand, has a multi detector system which covers 150 degree of scattering angle at once, as shown above. Using this multi detector system, one can obtain a hologram image by just rotating the sample without scattering angle scan. This drastically reduces the duration of one holography measurement, and thus makes the holography experiments feasible.

Since neutrons are sensitive to hydrogen positions, the neutron holography technique can observe local structures of hydrogens in materials. Though X-ray holography is of course an important probe to investigate of local structures of atoms, the accuracy of observations of light atoms is poorer than that of neutron holography. Thus, neutron holography will be an important tool for investigations of hydrogen, lithium, oxygen nitrogen, which play important roles in functional materials, such as hydrogen storage materials.

Moreover, the results on HERMES indicate that magnetic holography must be feasible. Since neutrons are sensitive to observe spin-spin correlations in magnets, magnetic holography will become a direct probe to observe local spin structures, while it is quite difficult or almost impossible to obtain accurate data on spin correlations by X-ray holography. The project aiming at realizing magnetic holography experiments on HERMES is now in progress in IMR.

For magnetic holography, polarized neutron holography must be most ambitious for novel material science. Since a polarized neutron beam includes only neutrons with the same spin direction, the polarized neutron technique is most sensitive to observe spin-spin correlations. Neutrons with the parallel and antiparallel spin to the magnetic moments in the sample give different and independent hologram images. Thus, this freedom on neutron spins will provide rich information of spin structure in magnets as well as enhancement of accuracy. Since the measurand of polarized neutron inverse holography will be intensity of \( \gamma \)-ray from the sample, \( \gamma \)-ray detection system must be installed to a polarized neutron spectrometer. By novel neutron holography experiments, we expect advances in some investigation fields: local structures of magnetic moments and hydrogens, structural physics investigations without models, structural investigations in materials which include neutron absorber elements, such as Sm, Gd, Cd. We are now preparing for preliminary experiments of polarized neutron holography in JRR3.
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