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Pendellösung interferometry by using pulsed neutrons

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ABSTRACT

Pendellösung interferometry is one of the technique for accurate determination of the structure factors of crystals. Observation method of Pendellösung fringes by using pulsed cold neutrons and the time-of-flight analysis were established. We measured the nuclear scattering length of silicon by the Pendellösung fringes with pulsed neutrons as $(4.125 \pm 0.003(\text{stat.}) \pm 0.028(\text{syst.}))$. This indicates the applicability of Pendellösung interferometry at high-intensity pulsed neutron facilities for various precision measurements.

1. Introduction

Pendellösung interferometry, which is based on the dynamical diffraction behavior in a perfect crystal, is one of the most accurate technique for determination of the structure factors of the crystals. Although measurements with the Pendellösung interference fringes require large scale of the perfect crystal, it has the advantage to extract the absolute value of the structure factor. Pendellösung interference fringes were explained theoretically by C. G. Darwin [1] and P. P. Ewald [2]. They were observed for the first time in 1959 by N. Kato and A. R. Lang [3]. Pendellösung fringes of neutrons were also observed in the late 1960's by C. G. Shull and so on [4–6].

In the case of symmetrical Laue geometry (Fig. 1(a)), when a neutron beam is injected into a perfect and thick crystal under the Bragg condition, the wave function in the crystal can be written as a superposition of the four components by the dynamical diffraction theory [7–12] as

$$\psi = \psi_0^1 + \psi_0^2 + \psi_g^1 + \psi_g^2, \tag{1}$$

where subscripts 0 and g represent the transmitted and the reflected wave, respectively, and subscripts 1 and 2 represent two Bloch wave functions. The wavenumbers of these Bloch waves are slightly different corresponding to the periodical nuclear potential in the crystal. In general, the four components are coherent and can interfere with each other. The radiation density is given by

$$|\psi|^2 \approx |\psi_0^1 + \psi_0^2|^2 + |\psi_g^1 + \psi_g^2|^2, \tag{2}$$

as the intensity of $(\psi_0^1 + \psi_0^2)^*(\psi_g^1 + \psi_g^2) + (\psi_0^1 + \psi_0^2)(\psi_g^1 + \psi_g^2)^*$ oscillates rapidly in the space that is so much smaller than the exit slit width to be smeared by averaging. The transmitted wave $|\psi_0^1 + \psi_0^2|^2$ and the reflected wave $|\psi_g^1 + \psi_g^2|^2$ generate a periodical structure of the radiation density in the crystal. This structure is known as Pendellösung interference fringes. Fig. 1(a) shows also the intensity distribution of neutrons on the end surface of the crystal, when neutrons is injected into the crystal through the narrow slit with the Bragg angle θ with respect to the crystallographic plane (hkl). The normalized distance $\Gamma \equiv x/(t\tan\theta)$ at the position x from the diffraction center along the end surface would be introduced, where t is the thickness of the crystal (see Fig. 1(b)). The intensity of the transmitted neutrons at the position Γ can be written as [10]

$$I_0(\Gamma)d\Gamma = {u_0}^2 \Delta_0 \tan\theta \frac{1-\Gamma}{(1+\Gamma)\sqrt{1-\Gamma^2}} \times \left[1 + \cos\left(\frac{\pi}{2} + \frac{4tF_{hkl}d_{hkl}}{V_c}\sqrt{1-\Gamma^2}\tan\theta\right)\right] d\Gamma, \tag{3}$$

where ${u_0}^2$ is the neutron density on the incident surface, F_{hkl} and d_{hkl} are the crystal structure factor and the spacing of the (hkl) crystallographic plane of the crystal, respectively, and V_c is the unit cell volume of the crystal. Δ_0 is the Pendellösung length, which is given by

$$\Delta_0 = \frac{V_c \pi \cos\theta}{\lambda F_{hkl}}, \tag{4}$$

where λ is the wavelength of the neutron. The neutron intensity through the exit slit for the transmitted beam, P_0, can be obtained by integration of Eq. (3) as

$$P_0 = \int_{-\Gamma_s}^{\Gamma_s} I_0(\Gamma)d\Gamma. \tag{5}$$

In the case of the symmetric and narrow slit with the width of $2\Gamma_s$ and $\Gamma_s \ll 1$, Eq. (5) can be expressed as

$$P_0 \simeq 2{u_0}^2 \Delta_0 \Gamma_s \tan\theta \left[1 + \cos\left(\frac{\pi}{2} + \frac{4tF_{hkl}d_{hkl}}{V_c}\tan\theta\right)\right]. \tag{6}$$

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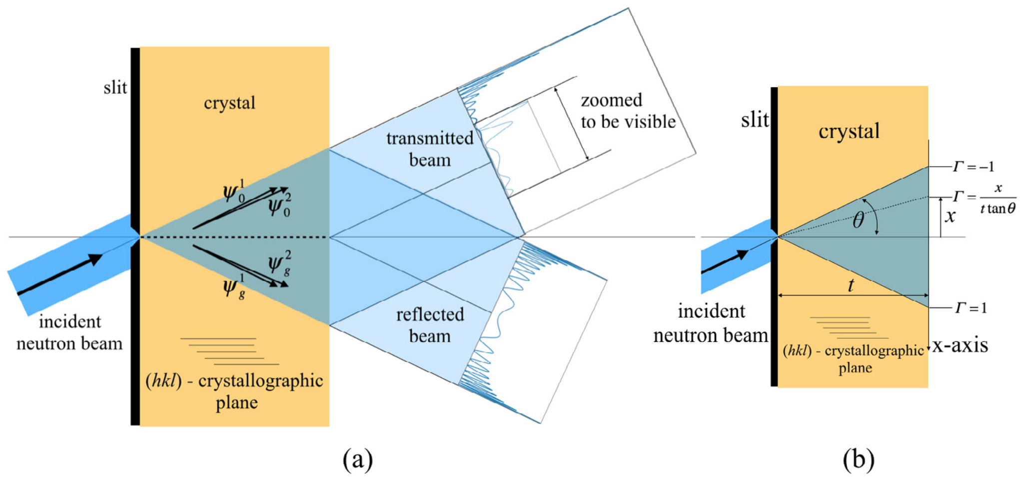

Fig. 1. (a) Neutron wave functions with symmetrical Laue geometry in a crystal can be written as a superposition of the four components by the dynamical diffraction theory. The incident neutrons propagate through the crystal spreading over a triangle zone (Borrmann fan) with a particular oscillating pattern due to the interference between the two Bloch waves in the transmitted and reflected beams, respectively. The curves on the right side represent the distributions of the emitted neutrons from the end surface of the crystal for the transmitted and reflected neutrons, respectively. In the transmitted direction, the intensity of the beam in the edge of the Borrmann fan is very strong, thus the central part is magnified to be visible. (b) Definition of the normalized position Γ on the end surface of the crystal.

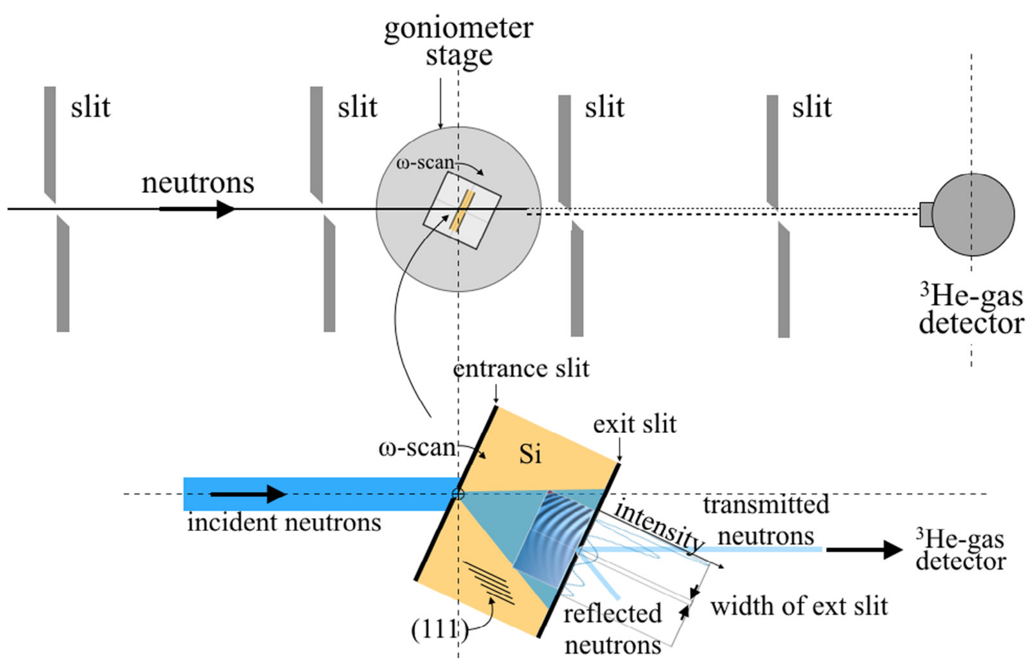

Fig. 2. Experimental setup to demonstrate Pendellösung fringes at the MLF BL17 in J-PARC (top view). The goniometer stage was scanned clockwise in the range of 25.0° – 26.5°. The real Bragg angle was obtained from the value of the TOF-position of the diffraction peak. The transmitted beam from the exit slit on the end surface of the crystal was shifted 1 mm downward in this figure and detected by the 3He-gas detector. The beam divergence was confined by the upstream two slits and entrance slit. Direct beam was cut off by the exit slit and the downstream two slits.

This periodic structure can be observed by varying θ [13]. This can also be measured by varying the thickness of the crystal [6,14]. The measurements of Pendellösung fringes enable the accurate determination of the crystal structure factor, and then it can be used to obtain the value of the coherent scattering length of the atom. Using this method, Shull and Oberteuffer obtained a nuclear scattering length of b_{nuclear} = (4.1491 ± 0.0010) fm for silicon [13]. These previous experiments were performed by using neutrons from reactors, therefore, monochromatic neutrons with the narrow energy-band was needed in order to satisfy the Bragg condition for a particular crystallographic plane. In this paper, we report the methodology of the observation of the Pendellösung fringes by using pulsed neutrons and the applicability of pulsed neutron beam to study fundamental physics by treating several crystallographic planes at the same time by using the time-of-flight analysis.

2. Experiment

We used a single silicon crystal with a width of 50 mm, height of 50 mm, and thickness of 2.8 mm, which was cut out from a float-zone ingot. The crystallographic plane (111) was used for demonstration. The surfaces of the crystal were mechanically polished and finally finished by chemical wet etching in order to remove the mosaic surface layer. The X-ray rocking curve exhibited a sharp peak with the width of less than 3 arcsec. The experiment using pulsed neutrons was performed at the beam line BL17 in the Materials and Life Science Experimental Facility (MLF) in J-PARC (Japan Proton Accelerator Research Complex). Fig. 2 shows the experimental setup. The crystal was sandwiched between the entrance cadmium slit and the exit cadmium slit which are 1.0 mm thick and have a 45° tapering. The width of both the entrance and exit