Holographic grating-mirrors for very cold neutrons

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The aim of our experiment was to demonstrate mirror-like behaviour of thick diffraction gratings, holographically recorded in nanoparticle-polymer composites for very cold neutrons. We have tested large-area free-standing film gratings that reached a reflectivity of 0.9 for $\lambda \sim 4.1$ nm.

An important task in the design of neutron-optical elements is structuring the neutron refractive-index of materials in an efficient way. For this purpose we utilize materials that are sensitive to light, combined with holographic techniques to produce diffraction gratings for neutron-optics. Due to the photo-neutron-refractive effect [1], nanoparticle-polymer composites exhibit a periodic neutron refractive-index pattern, arising from a light-induced redistribution of SiO₂ nanoparticles in the polymer matrix. Nanoparticle-polymer composites [2] offer advantages over polymer-dispersed liquid-crystals (H-PDLC) [3] or deuterated poly(methylmethacrylate) [4] for efficient tuning of the refractive-index modulation due to the possibility to choose the nanoparticle species. Recently, the experimental demonstration of a 50:50 beam splitter for cold and very cold neutrons (VCN) was achieved [5, 6] using such materials. The recorded grating thickness is - so far - limited to about 100 microns due to detrimental light-scattering during recording of the hologram. This obstacle has been overcome by increasing the effective grating thickness by tilting the grating around an axis parallel to the grating vector [7]. Thereby, the diffraction efficiency is increased because of the Pendellösung interference effect [8].

The main limitations of such gratings for neutron applications have been (*i*) the small sample size that limits also the beam size, especially for large tilt angles, (*ii*) intensity loss due to absorption/incoherent scattering by glass sample covers and the sample itself, which would pose a problem for wavelengths $\lambda > 5$ nm.

It was the main aim of our experiment, to demonstrate mirror-like reflectivity. We achieved this at large tilt angles ($\sim 70^{\circ}$), using large samples of 2 cm diameter. Therefore, the beam could be adjusted to a height of 5mm. For future experiments, the flexibility and reduced intensity loss of freestanding polymer-films (without glass covers) can be an advantage. Consequently, the second important result of our measurements is that, although the neutrons hit a large part of the sample area (about $0.5 \times 15 \text{ mm}^2$ for $\zeta = 70^\circ$), the grating-structure is homogeneous enough to exhibit mirrorlike diffraction efficiency. Neither does the removal of glass plates deteriorate the optical quality, nor did we observe deformation of the grating structure. Since there was a timeof-flight (TOF) system available during our beam time, we left the planned tests of a monochromator system using thin Si-wafers for future experiments.

In short, the sample preparation is as follows: The monomernanoparticle-mixture syrup is cast on a spacer-loaded,



Figure 1: Experimental setup for measuring mirror-like diffraction efficiency of holographic free-standing film-gratings.

silanized glass plate and is dried before covering it with another glass plate to obtain film samples [9]. A sinusoidal hologram with a grating spacing of the order of microns is recorded in the material by laser interferometry. Due to silanization, the glass plates can easily be removed using a razor blade, and we obtain somewhat flexible, 100 microns thin, free-standing film-gratings.

A SANS experiment to measure the diffraction efficiency of various samples was set up at PF2 VCN (see Fig. 1). The VCN beam, incident from the curved neutron guide, was reflected by a Ti/Ni-mirror so that a spectrum with intensity maximum at about 3.9 nm was obtained. Several collimation slits (Cd) provided for a beam cross section of $0.5 \times 5 \text{ mm}^2$ and horizontal/vertical angular divergence of no more than 1/3 mrad. The collimation distance was roughly 2.1 m. The sample was mounted on a rotation stage with 0.001° accuracy. The sample holder is constructed such that the grating can be tilted by an angle ζ about an axis parallel to the grating vector. The best result was achieved with a grating of $d \sim 100 \,\mu\text{m}$ thickness and a grating constant of $\Lambda = 0.5 \,\mu \mathrm{m}$ recorded in a SiO₂ nanoparticle-polymer composite at $\zeta = 70^{\circ}$. The diffraction efficiency at the Braggangle reached about 0.9 for $\lambda \sim 4.1$ nm. Some raw data are shown in Fig. 2. A more detailed discussion will be found in a dedicated publication [10]. Diffraction occurs clearly in the two-wave coupling (Bragg) regime, so that no intensity is lost to unwanted diffraction orders, as it would be the case for thin gratings.

The remaining problems in such experiment are:

- 1. The recording material was not deuterated, which again causes absorption/incoherent scattering.
- Monochromaticity: As measurements and simulations show, a very broad spectrum can cause broadening of the rocking curves. Therefore, the TOF-system is needed to observe mirror-features of the grating. Conse-



Figure 2: Left: 2D-detector matrix at off-Bragg position. All the intensity is transmitted to the 0^{th} diffraction-order spot. Right: 2D-detector matrix at the Bragg-angle. Here, almost all the intensity is diffracted to the $+1^{\text{st}}$ diffraction-order spot. The grating acts as a mirror with reflectivity 0.9.

quently, the biggest part of neutrons is lost to the chopper (with open-to-closed ratio of 3% in our case!).

The first problem can be solved by using deuterated monomer raw material for grating production, probably also in collaboration with the EMBL-ILL deuteration lab. However, as long as we work at $\lambda \sim 4$ nm with only one grating, intensity losses ($\sim 25\%$ of the total incident spectrum per grating) are tolerable. A test of a possible solution to the second point was already scheduled for this experiment but will, however, be realized in the near future instead: We want to test 0.2 mm thin Si wafers that reflect the longer wavelengths of the incident beam, transmitting a narrower wavelengthband to the sample.

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