Diamond-like carbon coatings for Ultracold Neutron applications

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Abstract

Diamond-like carbon (DLC) is a promising new wall coating material for use in applications with Ultracold Neutrons (UCN). It can potentially replace the toxic beryllium which has been widely used for the storage and transport of UCN.

Our aim is to produce DLC-coated neutron guide tubes of up to 1.2 m length with a Pulsed Laser Deposition (PLD) setup which is currently under construction. In order to find optimal process parameters and appropriate settings for the tube coating facility, we are carrying out a research program which includes the production and characterization of DLC films on small test samples.

The characterization of the test samples is done by Raman spectroscopy, X-ray Photoelectron Spectroscopy (XPS) and Neutron Reflectometry. We report on first results from the test samples with sp^3 fractions up to 55% and give an overview of the tube coating setup.
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1. Introduction

Ultracold Neutrons (UCN) have velocities up to 8 m/s (corresponding to kinetic energies below ~ 300 neV) and can be described as an ideal gas with a temperature of several milli-Kelvin [1]. At these low energies their trajectories in vacuum are affected by gravitation, by magnetic fields and by the strong interaction with nuclei. In order to describe the strong interaction, a pseudo potential, called the Fermi potential, can be defined for each material. Neutrons with kinetic energy below the Fermi potential of a material are reflected under any angle of incidence. From the Fermi potential V, the critical velocity

$$v_c = (2 \cdot V / m_n)^{1/2}$$

can be calculated, where m_n is the mass of the neutron. As the UCN energy spectrum usually follows a Maxwellian, the probability for finding a neutron is proportional to its kinetic energy and it is therefore of advantage to use a material with a high Fermi potential.

On each collision with the wall the UCN has a certain probability to be absorbed or inelastically scattered, i.e. to be lost. This loss probability is proportional to the loss coefficient η, which is material-dependent. Optimal materials are therefore materials with high Fermi potential and small loss coefficient. So far, beryllium ($V=252$ neV) has been widely used for that purpose, but as it is highly toxic there are efforts made to replace it.

One of the most promising replacement candidates is diamond-like carbon (DLC). Pure diamond with a sp^3 fraction of 1 has a Fermi potential of 304 neV which is consequently the theoretical limit for DLC. At the same time, the theoretical loss coefficient of diamond is one of the lowest of all known materials.

For UCN applications, the materials used should not contain large amounts of hydrogen as then the UCN are immediately upscattered on hydrogen atoms. The DLC coating should therefore be produced in a process which minimizes the amount of incorporated hydrogen. This can be realised using a Physical Vapour Deposition (PVD) method such as Pulsed Laser Deposition (PLD) [2]. For UCN applications, the coating thickness should be in the range of 100 to 300 nm.

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Recently, the performance of DLC for UCN applications was studied in detail [3]. It was shown that DLC is comparable to beryllium with respect to loss and depolarization upon wall collisions. The DLC coatings in this study were produced by PLD [4] and by vacuum arc deposition [5].

At PSI we are presently setting up a new, high-intensity source for UCN [6]. Spallation neutrons will be produced by the impact of 600 MeV protons from the PSI proton accelerator on a lead target. These neutrons are subsequently moderated to thermal energies in a heavy water volume and further slowed down to the UCN regime by 30 l of solid deuterium. The production of UCN will be performed during 8 s in intervals of 800 s. As the typical UCN experiments have filling times longer than the production pulse, the UCN have to be stored in between two production pulses. A prototype of such a storage volume, made out of DLC-coated stainless steel foils, has been recently tested with promising results [7].

In addition to the storage volume, we will also employ DLC-coated neutron guides. In order to produce these guide tubes, we are currently setting up a PLD facility, as presented in Section 4.

2. Coating of small test samples

We used an existing PLD setup at PSI to produce flat test samples of 10×10 mm^2 and 20×20 mm^2 size. This setup is described in [8]. We investigated the influence of different process parameters on the film quality, i.e. on the film density and the corresponding sp^3 fraction. The process parameters studied were the distance between target and substrate, the coating angle, the laser fluence and the vacuum conditions during deposition.

Spectroscopically pure graphite with a density of 2.21 g/cm^3 was used as target material. Most of the DLC films were deposited on standard silicon wafers; a few samples have also been produced with stainless steel substrates, in order to check for a dependence of the film density on the substrate material.

For the production of the test samples, an excimer laser (Lambdaphysik LPX105E, KrF, 248 nm) and a Nd:YAG-laser with frequency quadrupling (266 nm) were used. The samples were produced at pressures in the range of 10^{-5}–10^{-3} Pa.

In order to study the ionic composition (see e.g. Ref. [9]) of the ablation plume emission spectra of the plume were recorded with a spectrometer (ARC SpectraDrive).

After the deposition, the thickness of the deposited DLC films was measured with a profilometer (Dektak 8000).

3. Characterization results

We have recently developed a suitable characterization procedure for our test samples by conducting a comparative study of five different characterization methods for DLC samples [10]. Following this, the DLC films were measured with Raman Spectroscopy, X-ray Photoelectron Spectroscopy (XPS) and with Cold Neutron Reflectometry (only the 20×20 mm^2 samples).

The Raman measurements were performed on a Dilor LabRam spectrometer. The excitation light source was a HeNe laser at 632.8 nm with a power of 25 mW. A grating with 1800 l/mm was used. Following the procedure described in Ref. [10], we used a calibration curve to calculate the sp^3 fraction and the film density from the width of the Raman G band. The original calibration curve was extended to correctly evaluate the lower film densities of our test samples (compared to the reference samples used in [10]). The Raman G band and D band were both fitted with Gaussians.

Part of the samples was also measured with XPS. The measurements were performed on an ESCALAB 220i XL spectrometer using an Al Kα monochromatic X-ray source. For the fit parameters in the analysis, the same constraints as in [10] were used.

The samples with 20×20 mm^2 size were additionally measured with Cold Neutron Reflectometry, which is not a commonly used method to characterize DLC films but very suitable for DLC used in UCN applications. Cold neutrons have velocities above those of UCN, up to 1000 m/s. If the velocity component normal to the reflecting surface is smaller than the critical velocity, the neutron is reflected. This is the case for cold neutrons under glancing incidence. The reflectivity is measured as a function of the neutron velocity and the angle of incidence. The critical velocity can then be derived from the reflectivity curve.

All three methods show consistent results for the investigated test samples. Diamond fractions in the range of 20–55% were measured, depending on the different production process parameters used.

The dependence of the sp^3 fraction on the distance between target and substrate was investigated first. The results from a sample series, in which all process parameters except for the target–substrate distance were fixed, showed that the film density is constant for distances in the range of 2.5 cm–5.5 cm. However, as this series consisted of films with a low sp^3 fraction, we will repeat the series, choosing process parameters that will lead generally to a higher diamond fraction.

The second parameter studied was the laser fluence on the target. This was achieved by changing the voltage in case of the excimer laser and by using an attenuator with the Nd:YAG laser. Alternatively the laser fluence can also be changed via the position of the focussing lens, changing the spot size on the target. In general, we measured higher sp^3 fractions for higher laser fluences. Fig. 1 shows the XPS spectra of four samples, deposited with different laser fluences while all the other process parameters (distance between target and silicon substrate, pressure, laser wavelength) were kept constant. The values for the sp^3 fraction derived from the XPS spectra, following Ref. [11] and using the XPSPeak software [12], range from 22% for $1.3 \cdot 10^8$ W/cm^2 to 34% for $2.7 \cdot 10^8$ W/cm^2, which is close to the maximum laser fluence that was achieved and which may be marginal for high-density DLC films [13]. However, for our target the highest laser power in connection with an optimally focussed beam led to an undesired deposition of graphite particulates.