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Diamond mosaic crystals for neutron instrumentation: First experimental results

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ABSTRACT

Diamond single crystals were recently proposed as monochromators of unprecedented performance [1] (Freund, 2009). In the present paper we describe how diamond crystals with a suitable mosaic spread can be produced using a specific plasma CVD technique. Up to 2 mm thick samples with an average mosaic spread of 0.2° have been produced. We report on X- and gamma-ray characterisation checking the uniformity of the mosaic structure and present the results of a first study regarding the neutron reflection properties of this outstanding material. These promising results show that the diamond diffraction properties are not too far from the theoretical expectations. For example, 34% peak reflectivity has been obtained for a 1 mm thick crystal at 1 Å wavelength.

1. Introduction

Single crystal monochromators play a crucial role in neutron scattering instrumentation at continuous sources. Together with the source brightness and neutron beam properties, such as divergence, they determine the resolution in real and reciprocal space and the flux available for a neutron scattering experiment [2,3]. In most experiments, the full-width-at-half-maximum of the angular reflectivity distribution or rocking curve generated by neutron diffraction from an imperfect single crystal, commonly called neutron mosaic spread, should match the beam divergence that ranges typically from 0.2° to 1°. Then intensity and resolution are said to be optimised. Neutron monochromators are typically 20 cm long and 20 cm wide. They are composed of smaller mosaic crystal slabs that are often oriented to achieve single or double focusing [4,5].

In a recent publication [1] it has been shown theoretically that single crystal diamond would be the most efficient monochromator material for thermal neutron beams. It would outperform all other materials that are presently used or have been proposed such as copper, germanium and even beryllium. Thanks to the higher reflecting power of diamond, important flux gains could be achieved in the thermal and higher energy range for most neutron scattering instruments at continuous sources. Moreover, the use of diamond monochromators would lead to less γ-ray and neutron background intensities. The main obstacle for the application of single crystal diamond in neutron instrumentation has been the serious difficulty of producing large crystals with suitable mosaic spread at reasonable cost. Existing natural and synthetic diamond crystals about a cubic centimetre in size are nearly perfect and thus not suitable, even if they were affordable.

Chemical vapour deposition (CVD) provides an alternative concept for the synthesis of diamond crystals that is not restricted to the size limitation of natural stones or the traditional high pressure high temperature (HPHT) technique. Based on CVD methods, the production of hetero-epitaxial layers of diamond on various single crystal substrates has been reported quite recently. Among these, the most promising multilayer concept uses iridium/oxide buffer layers on single crystal silicon [6,7]. Thin diamond layers with a mosaic spread of 0.2–0.3° on areas of at least several square centimetres have been obtained. This technique has a real potential of scaling the substrate size up to several inches in diameter and several millimeters in thickness. Routine production of such big crystals with tailored mosaic spread would represent an absolute breakthrough.

Before embarking on a large scale research and development programme of fabricating diamond monochromators based on this novel technique, it was important to characterise thin, but readily available samples with respect to not only their defect structure but also their neutron diffraction properties. After successful completion of this first step, up to 2 mm thick

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specimens were produced. The mosaic structure with its spatial and angular uniformity was determined using X- and γ-ray diffraction and the neutron reflecting properties have been examined. Before presenting the deposition technique and the other experimental work, we briefly recall some relevant definitions concerning neutron monochromator crystals, see also [1] and the references given there.

2. Neutron reflection properties of mosaic crystals

2.1. Theoretical background

Monochromator crystals select a wavelength bandwidth $\Delta\lambda/\lambda$ out of a white incident neutron beam by Bragg diffraction at an angle θ_B according to the relation

$$\Delta\lambda/\lambda = [(\Delta d/d)^2 + (\alpha^2+\beta^2)\cot^2\theta_B]^{1/2} \quad (1)$$

The relative variation of the lattice spacing, $\Delta d/d$, is usually much smaller than the second term in the square brackets where α is the divergence of the incident beam in the scattering plane and β is called the *neutron mosaic spread*, which is the width of the neutron diffraction curve $r(\Delta)$, due to the angular misorientations of the mosaic blocks and Δ being the deviation from the exact Bragg angle. Gaussian shapes of the angular intensity distribution function and of the reflectivity curves are assumed. In general, intensity and resolution are optimised if α and β are about equal (*matching* condition), typically 0.2–0.8°. The rocking curve width, the peak and integrated reflectivity are principally determined by

- the intrinsic crystal properties, i.e. on one hand, the nuclear and structural parameters of the material and, on the other hand, its defect density and distribution giving rise to the mosaic spread; and
- the wavelength dependent neutron–material interaction processes, i.e. the attenuation processes by capture and scattering and the diffraction related phenomena, namely primary and secondary extinction.

The lattice tilts generated by the defect structure such as dislocations, grain boundaries, etc. are modelled by Darwin's mosaic block scheme assuming a normalised Gaussian angular distribution of the standard deviation η and centred at $\Delta=0$:

$$w(\Delta) = [\eta(2\pi)^{1/2}]^{-1}\exp(-\Delta^2/2\eta^2) \quad (2)$$

The FWHM, $\gamma=2.35\eta$, is called the *intrinsic mosaic spread*. It is always smaller than β which increases with wavelength as shown later. Because the divergence of a beam leaving a neutron guide tube also increases with wavelength, in this case the matching condition can be maintained to some extent when tuning the energy.

As mentioned above, variations of d due to strain or changes in the lattice constant can usually be neglected [8]. They are not considered by the model but can have an effect on the amount of primary extinction. On the other hand, the size of the mosaic blocks enters the theoretical treatment. The block size should be small enough so that primary extinction is insignificant, otherwise the reflectivity will drop. In the following we give a short summary of the theoretical background. More details of diffraction by mosaic crystals and monochromator materials properties have been given in Refs. [1,9–11].

The quality of a monochromator crystal is characterised by

- high scattering power inside the selected wavelength range;
- low scattering background and low contamination from higher orders; and
- low absorption (capture) cross-section and very small other attenuation factors (incoherent, parasitic, inelastic scattering).

A further important point is the availability of large single crystals with suitable mosaic structure (width of angular distribution of mosaic blocks, size of individual mosaic blocks, uniformity) at an acceptable cost level. The theory briefly recalled below is based on secondary extinction only and specific for the symmetric Bragg case where the beam enters and exits from the same face under the same (Bragg) angle. This is the most widely used geometry for neutron monochromators, because it gives always higher reflectivity than the transmission geometry [12]. Asymmetric reflections limit the energy tunability.

The first condition for high quality is met by high peak reflectivity, $r_p=r(\Delta=0)$, which is the ratio of reflected to incoming intensity for a given wavelength and mosaic spread. Both the peak and the integrated reflectivity (area under the rocking curve) increase monotonically with thickness t. For $t\to\infty$, r_p saturates at the maximum peak reflectivity, r_{pm}, which is given by

$$r_{pm} = a_0/(p_0+q_0) \quad (3)$$

where $a_0=w(\Delta=0)Q/\mu$, $p_0=1+a_0$ and $q_0=(1+2a_0)^{1/2}$.

The quantity $Q=(F_{hkl}e^{-W}/V_c)^2\lambda^3/(\sin 2\theta_B)$ is the scattering power per unit length, F_{hkl} is the structure factor and e^{-W} is the temperature dependent Debye–Waller factor with $W=(B_0+B_T)/(2d_{hkl})^2$. It depends on the Debye temperature, θ_D, and the atomic mass number, A; see the following section. V_c is the volume of the unit cell and d_{hkl} is the spacing of the diffracting lattice planes with Miller indices h, k and l. Inspection of Eq. (3) reveals that reflectivity is the higher, the bigger is a_0. This is equivalent to big Q and small μ, the latter being the attenuation factor per unit length, also called macroscopic attenuation cross-section. The intensity decreases along the beam path inside the crystal according to $I_T=I_0\exp\{-\mu t/\sin\theta_B\}$. Elastic coherent scattering competes with attenuation in the diffraction process. Attenuation is composed of neutron capture and by scattering processes such as incoherent and inelastic scatterings [13].

In a way similar to the reflectivity, the width of the reflectivity distribution also increases and the maximum width, β_m, for $t\to\infty$ is obtained as

$$\beta_m = 2\eta\left\{2\ln[a_0(2-r_{pm})^2/4r_{pm}]\right\}^{1/2} \quad (4)$$

To optimise the ratio of reflected neutrons to unwanted background and for practical reasons too, cost being one of them, it is useful to limit the thickness to a certain value where, say, $x=80\%$ of the maximum peak reflectivity is reached. It is therefore convenient to define such an *optimum thickness* t_x:

$$t_x = (\lambda/4d_{hkl}\mu q_0)\ln[(s_0+1)/(s_0-1)] \quad (5)$$

where $s_0=(1+p_0/q_0)/x-p_0/q_0$. The above equations are not valid for very small Bragg angles and for angles close to 90° where Q tends towards infinity. These extreme cases will not be considered here.

To obtain high reflectivity, the atoms composing a monochromator crystal should have high coherent scattering power and low incoherent and capture cross-sections. Heavy atoms and strong binding forces are favourable, because in this case neutron scattering is less affected by thermal vibrations. The latter are taken into account by the Debye–Waller factor and degrade the monochromator efficiency in two ways. First, the thermal diffuse scattering processes affect the coherent elastic scattering, and second, they increase the background intensity. If both capture processes and incoherent scattering intensity are small, inelastic scattering represents the main contribution to the total attenuation [13–16]. Moreover, background can become an important issue if the success of the experiment depends on *signal-to-noise* rather than flux. Details about attenuation and first experimental results for diamond single crystals will be published separately.