

# A NEW GENERATION OF X-RAY OPTICS BASED ON PYROLYTIC GRAPHITE\*

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## Abstract

Highly Oriented Pyrolytic Graphite (HOPG) is a mosaic crystal, which consists of a large number of small nearly perfect crystallites. The unique structure of HOPG crystals enables them to be highly efficient in diffraction in an energy range between 2 keV up to several 10 keV. The mosaicity of the crystal is responsible for the dramatic increase of integral reflectivity in comparison to perfect crystals. Furthermore thin HOPG crystal films can be easily bent and exhibit a very high thermal conductivity making them interesting for application in experiments with high average power x-ray sources. For application in x-ray spectroscopy the achievable spectral resolution of the crystal optics is of particular interest. Recently performed measurements with very low foil thickness have revealed a spectral resolution of  $E/\Delta E = 2900$  in (004)-reflection. This is by far the highest spectral resolution reported for Pyrolytic Graphite (PG) crystals. The integral reflectivity of these films is still comparable to that of ideal Ge crystals. In this work we present new results concerning the energy resolution, integral reflectivity and application of thin bent Graphite films.

## INTRODUCTION

Highly oriented pyrolytic graphite (HOPG) is an artificial graphite produced by thermal cracking of a hydrocarbon gas and deposition under low pressure and afterwards annealing the deposit under pressure [1]. In this way and after some further fabrication steps thin films of HOPG can be made with thickness of less than  $10 \mu\text{m}$ . These films can be easily bent by mounting them adhesively on a mould of any shape [2].

Sectional topographical diffraction measurements have shown an arrangement of the small crystallites with some  $\mu\text{m}$  in size to larger mosaic blocks with some  $100 \mu\text{m}$  in diameter and some  $10 \mu\text{m}$  in thickness [3]. The angular distribution of the crystallites, with plane orientations off to the normal axis to the crystal surface, is called mosaic spread. The mosaic spread between the crystallites is much smaller than that between the mosaic blocks. The mosaic blocks again show a correlation in orientation and form larger crystal domains with some mm in size.

Because of its high thermal conductivity  $\kappa$  and low linear thermal expansion coefficient  $\alpha$  in basal plane (cp. Ta-

ble 1), which both are similar to those of diamond crystals, PG crystals could be interesting for many applications with regard to x-ray sources delivering high average x-ray power. Also the absorption thickness  $t_{abs}$  of HOPG is similar to that of diamond. Consequently less heat is absorbed in comparison to other crystals.

The diffraction properties influencing the energy resolu-

Table 1: Thermal properties and absorption thickness at 8 keV for different crystals.

Material (hkl)	$\kappa$ [W/cm K]	$\alpha$ [1/K x 10 <sup>-6</sup> ]	$t_{abs}$ [ $\mu\text{m}$ ]
Diamond(111)	21	0.8	250
Si(111)	1.25	2.33	8.7
Ge(111)	0.58	5.9	2.94
HOPG(002)			239
(parallel (002))	17	1	
(perpendicular)	8	20	
HOPG(004)			477

tion of PG films are determined by the mosaicity and the intrinsic width of Bragg reflection. Latter refers to the diffraction properties of the small crystallites and is called the Darwin width for nearly perfect crystals. It results from particle size and/or strain broadening. Mosaicity makes it possible that even for a fixed angle of incidence to the crystal surface, an energetic distribution of photons can be reflected, because each photon of this energetic distribution can find a crystallite plane at the right Bragg angle. The width of this energetic distribution depends on the mosaic spread. The mosaicity is also responsible for the dramatic increase of integrated reflectivity in comparison to perfect crystals in an energy range between 2 keV and several 10 keV [4]. The so-called mosaic-focusing, as shown in Fig. 1, which occurs in a 1:1 magnification geometry enhances further the intensity in the image plane.

In contrast to ideal crystals, in mosaic crystals the photon has to penetrate deeper into the crystal, before it finds a crystallite aligned well, from which it can be reflected. That means that the effective depth, from which diffraction in mosaic crystals occurs, is much larger compared to ideal crystals. Therefore the energy resolution can be strongly affected by the thickness of the Graphite films. Recently we have shown that very thin Graphite films can reveal energy resolutions and integral reflectivity comparable to Ge(111) [5].

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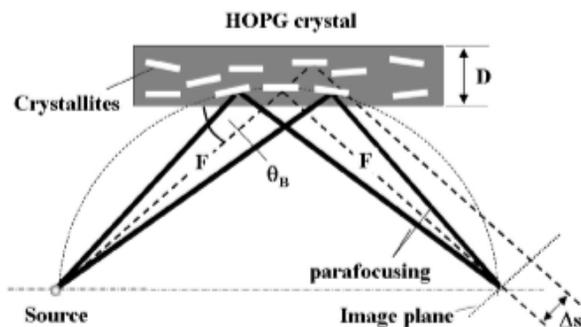


Figure 1: Diffraction properties of HOPG.

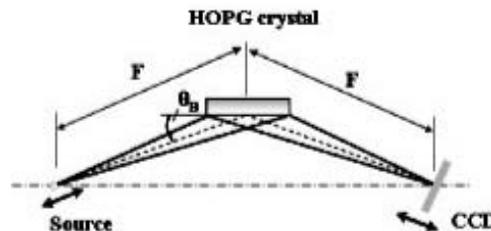


Figure 2: Experimental setup.

Because PG can be treated as an agglomeration of very small crystallites less strain is induced by bending the films and therefore no drastically changes of the diffraction properties determining the spectral energy resolution are expected and were measured up to now.

## EXPERIMENTAL AND RESULTS

### Samples

Two kind of HOPG crystals manufactured by Optigraph GmbH were investigated. Firstly, the well known "Highly Oriented Pyrolytic Graphite" (HOPG) crystal and secondly a new kind of HOPG crystal. This new material was already presented in [5]. Because the diffraction properties of the new material differ considerably from the well known HOPG it will be named by Optigraph henceforth "Highly Annealed Pyrolytic Graphite" (HAPG) to distinguish it from the commonly HOPG. This new kind of PG crystal is optimized for the application in x-ray spectroscopy by a modified fabrication process. To get a comprehensive overview over the relationship between energy resolution and thickness in flat and bent geometry, crystals with different thickness were measured in (004)-reflection in both geometries. To realize precise variations in thickness the PG films were fabricated by stacking thin 10 - 15  $\mu\text{m}$  thick PG films. The influence of the stacking on the energy resolution was investigated by measuring also thicker PG films consisting of only one sheet. The flat crystals were mounted on polished glass plates and the bent crystals were mounted on a cylindrical polished lens with radius 150 mm. The thickness of the investigated films was determined with an accuracy of  $\pm 5 \mu\text{m}$  using a micrometer screw before mounting them on the moulds. It must be noted, that the measured thickness is an averaged value. That means, that the thickness can vary slightly for different sites of the crystal.

### Experimental setup

The experimental setup is shown in Fig. 2. It consists of the HOPG crystals, an x-ray tube and a CCD camera.

The x-ray source, which was used for the measurements, is a low power microfocus x-ray tube (IfG) with a source diameter of about 50  $\mu\text{m}$ . Measurements were performed with the  $\text{Cu K}_\alpha$  emission of a Cu anode at 8 keV. The spec-

tra were collected with a 16-bit deep depletion CCD camera (Roper Scientific model PI-LCX 1300) with a quantum efficiency of about 50% at 8 keV. A thin (250  $\mu\text{m}$ ) Be window in front of the deep depletion CCD was used for vacuum sealing of the camera, so that a deep cooling (down to  $-50 \text{ }^\circ\text{C}$ ) of the CCD was possible. The distance between source and crystal and between crystal and detector was  $F = 400 \text{ mm}$  in each measurement. All measurements were carried out with the detector plane oriented perpendicularly to the reflected x-ray beam.

### Results

In Fig. 3 selected images of the reflected  $\text{Cu K}_\alpha$  emission are presented for both investigated PG crystals. In the upper image a 15  $\mu\text{m}$  HAPG and in the lower image a 10  $\mu\text{m}$  thick HOPG crystal in flat geometry is shown. As can be seen from the cross sections of the images in

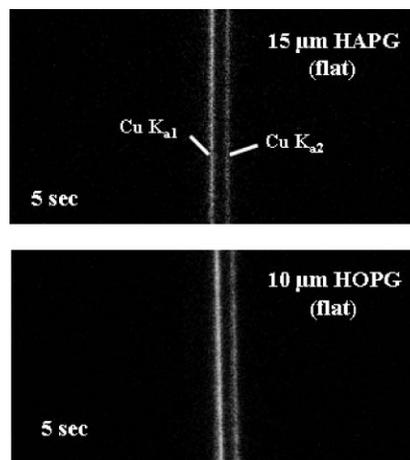


Figure 3: Recorded images of the reflected  $\text{Cu K}_\alpha$  emission from a 15  $\mu\text{m}$  thick HAPG and a 10  $\mu\text{m}$  thick HOPG.

Fig. 4 the measured energy resolution of the flat HAPG crystal is better than that of the HOPG crystal with nearly same thickness. The energy resolution was determined by a convolution procedure as described in [5]. From the convolution procedure an energy resolution  $E/\Delta E$  of 4100 was

found for the 15  $\mu\text{m}$  flat HAPG and 3500 for the 10  $\mu\text{m}$  flat HOPG. The reflection broadening due to crystal thickness, which is in the range of a single mosaic block, cannot explain the difference in energy resolution between both crystals. Therefore, the differences in energy resolution are more likely a broadening due to the flat geometry of the crystals. Latter gives rise to a focusing error in the image plane, because the alignment of the mosaic blocks in the basal plane deviates from a Rowland circle. These so-called flat focusing error increases with mosaicity (cp. [6]). Hence for the HAPG lower mosaic spread can be expected. As mentioned above the 15  $\mu\text{m}$  HAPG crystals are compa-

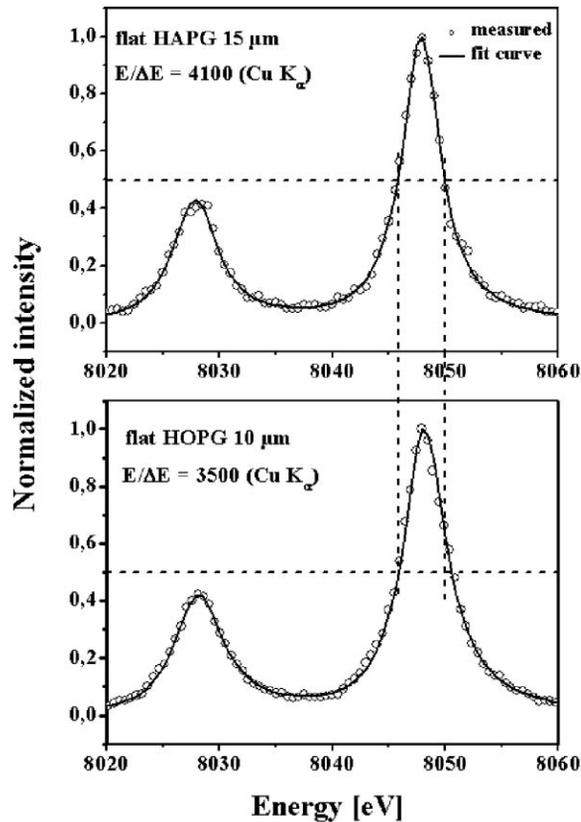


Figure 4: Cross sections of the recorded images in Fig. 3. The circles are measured points and the lines represents the fit curves of the convolution procedure.

table to the 15  $\mu\text{m}$  thick crystal which was presented in [5]. The energy resolution in Fig. 4 is higher because the distance  $F$  is larger. Taking the values for the energy resolution from [5] and comparing these values with the measured energy resolution at 400 mm distance a nearly linear increase of energy resolution over distance is observed.

As already reported in [5] the integral reflectivity of the 15  $\mu\text{m}$  thick HAPG in (004)-reflection is similar to that of Ge(111) crystals. For the 10  $\mu\text{m}$  HOPG the integral count rate on the CCD over the Cu  $K_{\alpha}$  emission is by factor two higher than that of the 15  $\mu\text{m}$  HAPG.

The energetic reflection bandwidth of these thin 15  $\mu\text{m}$

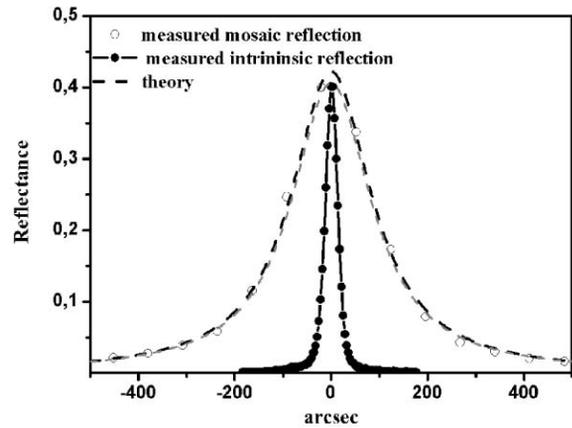


Figure 5: Measured rocking curves of a 15  $\mu\text{m}$  thick HAPG crystal in (002)-reflection. The measurements of the rocking curves were performed using a triple-crystal diffractometer (BESSY).

HAPG films can be determined by rocking curve measurements. The measurements of the rocking curves were performed in (002)-reflection using a triple-crystal diffractometer at BESSY. In Fig. 5 the mosaic reflection and the intrinsic width of reflection of HAPG(002) are shown. The rocking curve width (FWHM) is 0,056  $^{\circ}$  for the mosaic reflection and 27 arcsec for the intrinsic reflection. From the mosaic reflection the mosaic spread can be determined. To determine the mosaic spread from the measured rocking curves a reflectivity formula [7] derived from Zachariassen's treatment was used [8]. The best fit was obtained with a Lorentzian distribution and a mosaicity of 0.042  $^{\circ}$ . From the measured mosaic spread the energetic bandwidth can be calculated in both reflection orders.

To enhance the reflectivity the thickness of the HAPG

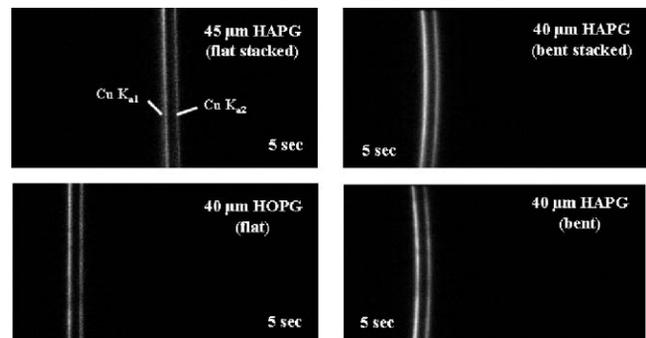


Figure 6: Recorded images of the reflected Cu  $K_{\alpha}$  emission from a stacked 45  $\mu\text{m}$  thick flat HAPG (upper left image), a 40  $\mu\text{m}$  bent stacked HAPG (upper right image) and a 40  $\mu\text{m}$  single sheet HAPG in flat (left lower image) and bent (right lower image) geometry.

films was increased. In Fig. 6 the images of a 40  $\mu\text{m}$  thick

HAPG single sheet and a stacked HAPG of same thickness are shown. Due to thickness broadening the energy resolution drops down to 3300 for both sheets. In contrast the integral count rate increases by a factor 4 for both 40  $\mu\text{m}$  PG films in comparison to the 15  $\mu\text{m}$  thick film. Also shown in Fig. 6 are bent HAPG crystals of same thickness. Bending these 40  $\mu\text{m}$  thick HAPG crystals revealed fully different energy resolutions. While bending increases the energy resolution for the single sheet of HAPG as shown in Fig. 7, for the stacked bent HAPG the energy resolution drops down to 2300 (not shown). This result was surprisingly and indicates maybe changes in the mosaic structure by bending. In the case of the stacked HAPG an interaction between different layer has to be taken into account.

Finally, to enhance further the integral reflectivity accom-

## CONCLUSION

High energy resolution can be obtained in (004)-reflection using very thin PG crystals. These high energy resolutions are accompanied with high integral reflectivity. As shown, bending single sheets of these very thin crystals seems not to decrease the energy resolutions.

Because of its good thermal properties PG could be interesting for many different applications based on high averaged x-ray sources, as e.g. the Free Electron Laser. A very promising application for PG crystals could be the single shot x-ray spectroscopy in an energy range between several keV up to several 10 keV. This can be done with PG because the mosaic spread enables to record a broad energy range with high energy resolution in a single shot.

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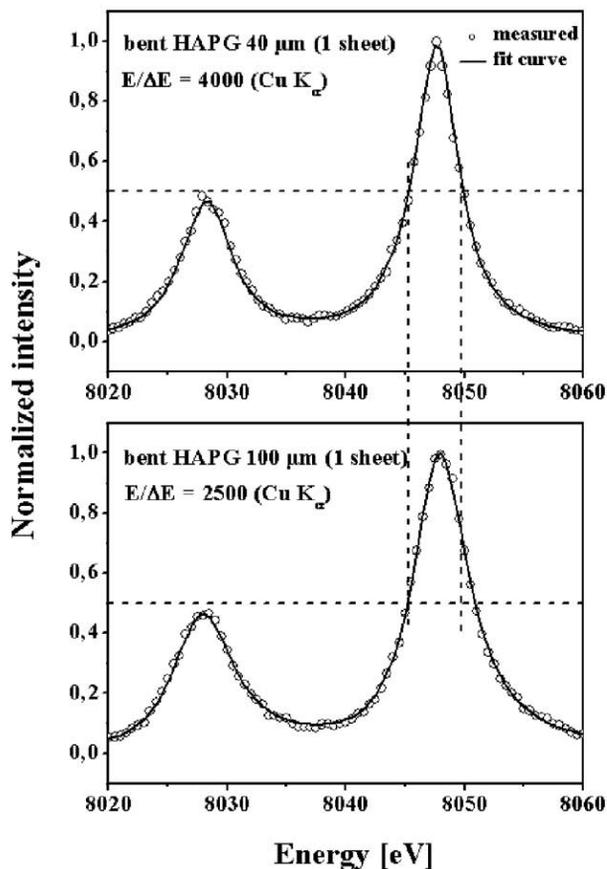


Figure 7: Cross sections of the recorded images in Fig. 6. The circles are measured points and the lines represents the fit curves of the convolution procedure.

panied with high energy resolution a 100  $\mu\text{m}$  thick single HAPG sheet was investigated. The integral count rate increases further by a factor 2 and the energy resolution drops down to 2500 as can be seen in Fig. 7.