Implementing X-Ray diffraction/scattering tomography in the lab

Real materials often exhibit a complex and heterogeneous organization. Their distinctive functionalities are linked to their specific structure at the atomic scale as observed in alloys, cements but also in teeth, bones or archaeological materials. We have developed a method combining the diffraction and scattering of X-Rays with computed tomography. The diffraction/scattering information is used to detect, identify and image the various crystalline/amorphous phases in heterogeneous samples. This non-destructive analysis technique, first developed on synchrotron radiation facilities, can now be implemented at medium resolution on ordinary laboratory X-Ray diffraction equipment.

Tomography (from the Greek "tomos" = "slice") is a method for generating virtual images of slices through an object. In Diffraction and Scattering Computed Tomography (DSCT), the object is mounted in a very narrow beam of monochromatic X-Rays, and the diffraction and scattering intensities are recorded with a 2-Dimensional (*i.e.* multi-pixel) detector, see Fig 1. This is done with the object oriented at a large number N_ω of rotational positions ω, for each of a similarly large number N_y of horizontal positions *y*. From this very large mass of data, one can compute an image of the slice that contains structural information: chemical composition, atomic order (crystalline or amorphous, type of phase), grain size, etc. If desired, the object can then be translated vertically in steps to build a 3D representation as a stack of slices. DSCT also increases sensitivity to weak X-Ray signals coming from minor phases.

To implement DSCT in-laboratory, with spatial resolution ~200 microns, we adapted a multi-circle goniometer normally dedicated to single-crystal diffraction measurements and we added the translational and rotational stepping motions required for tomographic data acquisition. To acquire one slice, we record N_y x N_ω (N~100) diffraction images on a multipixel 2D detector. The various, randomly oriented, lattice-planes (*hkl*) in polycrystalline material diffract the incident X-Ray beam through their specific Bragg deviation angles 2Θ, giving the concentric rings in Fig. 1. Any highly disordered or amorphous materials scatter diffusely.

As illustration, we discuss our analysis of a carbon sample resulting from the phase transformation of Carbon 60 (C_{60} "fullerene" molecules) into a mixture of a crystalline rhombohedral polymer (C60R) and disordered graphite by high pressure (5 GPa) at 1100 K. Each of the N_y x N_ω 2D diffraction/scattering patterns is integrated azimuthally around its centre to convert it into a 1D pattern, *i.e.* diffraction/scattering intensity vs the deviation angle 2Θ, like that of Fig. 2a where we have labelled strong Bragg

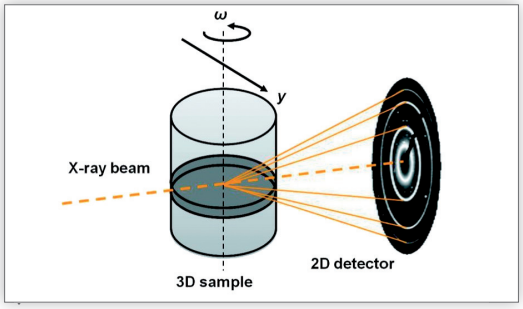

Fig. 1: *Geometry for X-Ray diffraction and scattering tomography analysis. To analyse one slice, the sample is translated in steps along y and rotated 180 deg. through angle ω.*

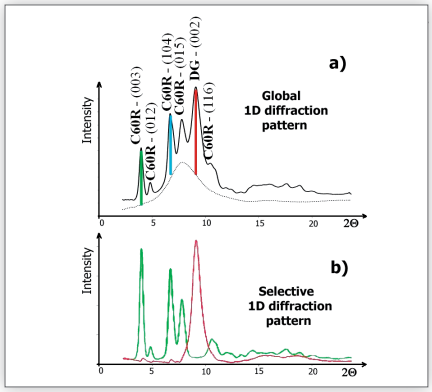

Fig. 2: (a) *A global 1D diffraction pattern for a slice through the transformed carbon sample shows Bragg diffraction peaks for lattice planes of its Rhombohedral C60 and disordered textured graphite (DG) components.*

(b) *Reverse analysis yields 1D-diffraction patterns selective for (in green) Rhombohedral C60 and (in red) disordered graphite.*

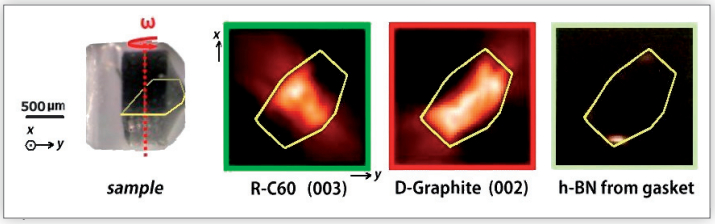

Fig. 3: At left: *The transformed carbon sample within its glass capillary container.*
At right: *Three phase-selective virtual slices as computed from the data for specific Bragg angles corresponding to (i) the (003) planes of the rhombohedral C60 component, (ii) the (002) planes of disordered textured graphite, and (iii) an unanticipated hexagonal-BN impurity. (The sample shape is outlined in yellow.)*

diffraction peaks for C60R and disordered graphite. From the complete set of N_y x N_ω 1D diffraction patterns, we can compute virtual slices that image the intensity scattered at a specific Bragg angle, versus *x* and *y* at height *z* in the sample. Additionally a reverse analysis of the transformed data can be performed in order to extract pure single-phase diffraction patterns (*e.g.* Fig. 2b) for all the phases within the heterogeneous sample, including unexpected components.

For our carbon sample (Fig. 3), we evidenced (i) rhombohedral C60 domains in the middle and lower part of the sample and (ii) disordered graphite domains in the external and top part of the sample. Interestingly (iii) a hexagonal Boron Nitride (h-BN) impurity phase coming from the high pressure gasket was also localized.

This work demonstrates that X-Ray diffraction/scattering tomography can be performed in the laboratory, with all the advantages of easy and instant access that this provides, even if the spatial resolution presently attainable (~100 microns) is very much lower than that obtainable at a Synchrotron radiation facility. Our technique will be particularly interesting for samples containing crystallites and/or heterogeneities a few hundred microns in size. It is suited for numerous practical applications like cements, alloys, catalysts, bones and paint cross-sections.

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FURTHER READING

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