Unfolding a two-dimensional powder diffraction image: conformal mapping

Antonio Cervellino,^a Cinzia Giannini,^b Antonietta Guagliardi^b and Massimo Ladisa^{b,c,*}

^aLaboratory for Neutron Scattering, ETH Zurich and PSI Villigen, CH-5232 Villigen PSI, Switzerland,

^bIstituto di Cristallografia (IC-CNR), Via G. Amendola 122/O, I-70126 Bari, Italy, and ^cInstitut Néel (CNRS), 25 Avenue des Martyrs, BP 166, 38042 Grenoble, France. Correspondence e-mail: massimo.ladisa@ic.cnr.it

A new procedure aimed at unfolding a two-dimensional powder diffraction image into both a one-dimensional azimuthal and a radial scan is presented. In this approach, the sample-to-detector distance is the only parameter that must be adjusted in a separate step by using a standard sample. The technique consists of three steps: tracking the beam centre as the local maximum of the self-convolution of the original two-dimensional map, detector tilt and rotation determination by an intensity-tensor diagonalization, and azimuthal/radial intensity integration by a conformal mapping of the original two-dimensional powder diffraction image. The X-ray powder diffraction (XRPD) intensity profile of the NIST Si 640c standard sample is used to test the performance. The results show the robustness of the method and its capability of efficiently tagging the pixels in a two-dimensional readout system by matching the ideal geometry of the detector to the real beam-sample-detector frame. The technique is a fast, versatile and user-friendly tool for the simultaneous analysis of both azimuthal and radial spectra of two-dimensional XRPD images.

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1. Introduction

Complex hierarchic biomaterials (bone, collagen, wood *etc.*) and template-controlled novel materials are examples of complex cases normally investigated with small-angle X-ray scattering (SAXS) and wide-angle X-ray scattering (WAXS) techniques (two-dimensional powder diffraction¹), which need high-throughput online data analysis and visualization (Paris & Riekel, 2005). In experiments related to bone-tissue engineering studies with porous bioceramic scaffolds, the local interaction between the newly formed mineral crystals and the biomaterial has been investigated by means of microdiffraction with submicrometre spatial resolution (Mastrogiovanni *et al.*, 2007; Guagliardi *et al.*, 2007). Moreover, the combination of the above-mentioned techniques (namely SAXS–WAXS) with the spatial resolution enables the analysis of the orientation of crystallographic axes of grains as well as the orientation of the mineral crystals and microfibrils of the engineered tissue with respect to the scaffold (Cedola *et al.*, 2006, 2007). Following the growth model of the natural tissue is crucial in order to check the grade of adhesion of the newly formed tissue within the scaffold. Thus a combined radial/azimuthal analysis characterizes both the mineralization

intensity and the orientation degree of the new material. With this aim, a high-throughput online automatic data-folding procedure for disentangling both radial and azimuthal patterns out of two-dimensional images is essential, provided that (i) it is user independent and (ii) it can be applied to a set of up to several thousand images. Indeed, this type of experiment produces large data sets. This is no longer a problem so far as data-collection time is concerned, as long as pixel area detectors (such as CCDs) are used (Takeda *et al.*, 2005; Hanley & Denton, 2005; Hammersley *et al.*, 1997; Muchmore, 1999; Phillips *et al.*, 2002; Suzuki *et al.*, 1999). On the other hand, besides the high computational cost, processing a large number of data sets can lead to the overlooking of or misinterpretation of some features if an automatic procedure is not implemented. In most of the widely used procedures (see, for instance, Hammersley *et al.*, 1997; Rajiv *et al.*, 2007; Hinrichsen *et al.*, 2007; Rodriguez-Navarro, 2006), folding requires a detector calibration step performed by means of a standard sample. This calibration allows one to find all of the geometrical parameters necessary to extract a one-dimensional profile from a two-dimensional image. In the present paper a new automatic procedure aimed at disentangling both radial (folding) and azimuthal patterns out of a two-dimensional powder diffraction image is presented. This procedure allows the user to set all geometrical parameters, except for the detector-to-sample distance, without any standard sample (Cervellino *et al.*, 2006).

¹ In this context 'two-dimensional' refers to the data array structure.

2. The method

The ideal X-ray powder diffraction (XRPD) geometry is cylindrical about the beam-sample axis. The diffraction cone with an opening angle 2θ and the apex on the sample reaches the imaging plate (a CCD detector, for instance). Ideally the imaging plate would be orthogonal to the beam-sample axis and it would collect a series of circles with radii corresponding to different momentum transfer: the larger the radius the higher the momentum transfer (*i.e.* the resolution). Unfortunately the imaging plate axis rarely fits the ideal geometry, and the circles become ellipses with an eccentricity ε and a rotation φ_0 parameterizing the tilt and rotation of the detector frame. Furthermore, the beam rarely reaches the geometrical centre of the plate and a global shift of the rings is needed to perform a correct two-dimensional folding. The problem reduces to the nonlinear fit of the parameters appearing in the Cartesian equation of an ellipse (Cervellino *et al.*, 2006).

In this paper we tackle the problem from a different point of view. We adopt neither a fitting procedure nor a statistical/dynamical approach to extract the parameters: we shall pursue a fully deterministic calculation scheme. In our opinion, the latter will be computationally less demanding and more straightforward than the former schemes.

We claim that all the parameters necessary to the folding procedure can be obtained independently of the detector-to-sample distance calibration. Let us suppose that the detector-to-sample distance is known exactly. The two-dimensional folding procedure presented here will allow us to determine the geometrical parameters, namely eccentricity, rotation and centre coordinates, without any need of a standard sample. The procedure is accomplished *via* three steps: beam-centroid determination, tilt/rotation angle fitting and radial intensity integration.

2.1. Beam-centroid determination

This task is carried out by calculating the local maximum of the self-convolution of the original two-dimensional XRPD image Φ . The rationale of this procedure can be understood as follows. If $\Phi = \Phi(x, y)$ is a function defined on the real plane $\mathbb{R}^2$, the self-convolution $\Phi \star \Phi(x, y)$ is defined as follows:

$$\begin{aligned} \Phi \star \Phi(x, y) &= \int_{\mathbb{R}^2} \int_{\mathbb{R}^2} dx' dy' \Phi(x - x', y - y') \Phi(x', y') \\ &= \int_{\mathbb{R}^2} \int_{\mathbb{R}^2} dk_x dk_y \exp[i(k_x x + k_y y)] |\tilde{\Phi}(k_x, k_y)|^2, \end{aligned} \quad (1)$$

where $\tilde{\Phi}(k_x, k_y)$ is the Fourier transform of $\Phi(x, y)$. Hereafter, we shall assume that the symmetry centre is placed at the origin of the plane. Moreover, since the determination of ε/φ_0 is a fine tuning with respect to the beam-centroid determination (Cervellino *et al.*, 2006), we shall also assume that Φ depends on $(x^2 + y^2)^{1/2}$ only. Extending the result to the general case is straightforward. Thus in polar coordinates the definition in (1) reads

$$\Phi \star \Phi(\rho, \theta) = \int_{\mathbb{R}^+} d\rho' \rho' \Phi(\rho') \int_{[0, 2\pi]} d\theta' \Phi(X), \quad (2)$$

where X is defined as $[\rho^2 + \rho'^2 - 2\rho\rho' \cos(\theta - \theta')]^{1/2}$, $\rho^{(i)}$ as $[x^{(i)2} + y^{(i)2}]^{1/2}$ and $\tan \theta^{(i)}$ as $y^{(i)}/x^{(i)}$.

The conditions for the origin on the plane $\mathbb{R}^2$ to be the local maximum for $\Phi \star \Phi$ are $\partial_\rho \Phi \star \Phi(0, \theta) = 0$ and $\partial_\rho^2 \Phi \star \Phi(0, \theta) < 0$, while the angular dependence can be neglected because of the central symmetry of Φ . Indeed, since

$$\begin{aligned} \partial_\rho \Phi \star \Phi(\rho, \theta) &= \int_{\mathbb{R}^+} d\rho' \rho' \Phi(\rho') \\ &\quad \times \int_{[0, 2\pi]} d\theta' \frac{\rho - \rho' \cos(\theta - \theta')}{X} \partial_\xi \Phi(X), \\ \partial_\rho^2 \Phi \star \Phi(\rho, \theta) &= \int_{\mathbb{R}^+} d\rho' \rho' \Phi(\rho') \int_{[0, 2\pi]} d\theta' \left\{ \frac{\partial_\xi \Phi(X)}{X} \right. \\ &\quad \left. + \left[\frac{\rho - \rho' \cos(\theta - \theta')}{X} \right]^2 \left[\partial_\xi^2 \Phi(X) - \frac{\partial_\xi \Phi(X)}{X} \right] \right\}, \end{aligned} \quad (3)$$

it immediately follows that $\partial_\rho \Phi \star \Phi(0, \theta) \propto \int_{[0, 2\pi]} d\theta' \cos(\theta - \theta') \rightarrow 0$, while $\partial_\rho^2 \Phi \star \Phi(0, \theta) \propto -\int_{\mathbb{R}^+} d\rho' \rho' [\partial_\rho \Phi(\rho')]^2$, which is negative whatever value the angle θ assumes. Thus the origin on the plane $\mathbb{R}^2$ is a symmetry centre for Φ and a symmetric local maximum for $\Phi \star \Phi$; moreover, since $\Phi \star \Phi(x, y) \leq \Phi \star \Phi(0, 0) \forall (x, y) \in \mathbb{R}^2$ [see equation (1)], the origin on the plane $\mathbb{R}^2$ is also a global maximum for $\Phi \star \Phi$.

The correct positioning of the beam centre onto the imaging plate is shown in Figs. 1(a)–1(c), together with the self-convolution of the original two-dimensional XRPD map and its maximum.

Figure 1
(a) Original XRPD pattern. (b) The local maximum of the original XRPD pattern self-convolution. (c) Original XRPD pattern together with the beam centre corresponding to the local maximum of its self-convolution. (d) Original XRPD pattern together with the beam centre and the inertia tensor principal axes.

2.2. Detector tilt and rotation determination

Once the beam centre has been positioned, the computation of the eccentricity ε and rotation φ_0 is straightforward; in fact the two-dimensional XRPD image Φ can be regarded as a form of planar mass density (indeed, both quantities, *i.e.* mass density and X-ray intensity recorded on the image plate, are positive by definition); we therefore compute an inertia tensor $\mathcal{I}$ and we diagonalize it. The Cartesian tensor $\mathcal{I}$ is defined as follows:

$$\mathcal{I} = \int \int_{\mathbb{R}^2} dx dy \Phi(x, y) \begin{pmatrix} y^2 & -xy \\ -xy & x^2 \end{pmatrix}, \quad \mathcal{I} = \mathcal{I}^\dagger. \quad (4)$$

Hereafter we shall assume that the function Φ is uniform along the same ellipse (as ideally expected for randomly oriented powders), *i.e.* $\Phi(a) = \text{constant}$, where a is the major semi-axis of a given ellipse, and we shall neglect the azimuthal dependence φ ; we define $x + iy = a \exp(i\varphi) [1 + \varepsilon \sin(\varphi - \varphi_0)]^{-1/2}$ as the local isomorphism between $\mathbb{R}^2$ and the complex plane $\mathbb{C}$ in the elliptical coordinates frame. These assumptions are general and they will not affect our results. In

such a frame the tensor $\mathcal{I}$ factors out the radial and the azimuthal dependence, *i.e.* $\mathcal{I} = \mathcal{I}_a \times \mathcal{I}_\varphi$, where the azimuthal and radial components are defined as

$$\begin{aligned} \mathcal{I}_a &= \int_{\mathbb{R}^+} da a^3 \Phi(a), \\ \mathcal{I}_\varphi &= \int_{[0, 2\pi]} \frac{d\varphi}{[1 + \varepsilon \sin(\varphi - \varphi_0)]^2} \begin{pmatrix} \mathcal{S}^2 & -\mathcal{S}\mathcal{C} \\ -\mathcal{S}\mathcal{C} & \mathcal{C}^2 \end{pmatrix} \\ &= \frac{\pi}{(1 + \varepsilon)^{1/2}} \left[\mathbb{I} - \frac{\varepsilon}{1 + \varepsilon} \begin{pmatrix} \mathcal{C}_0^2 & -\mathcal{S}_0\mathcal{C}_0 \\ -\mathcal{S}_0\mathcal{C}_0 & \mathcal{S}_0^2 \end{pmatrix} \right], \end{aligned} \quad (5)$$

with $\mathcal{S}_{(0)}$ and $\mathcal{C}_{(0)}$ denoting $\sin \varphi_{(0)}$ and $\cos \varphi_{(0)}$, respectively, and where $\mathbb{I}$ is the identity matrix. Equation (5) clearly shows that, while the radial integral $\mathcal{I}_a$ is a constant, the azimuthal integral $\mathcal{I}_\varphi$ carries the relevant information for the calculation of ε and φ_0 . Indeed, the latter are promptly obtained by diagonalizing the azimuthal tensor $\mathcal{I}_\varphi$, yielding $\varepsilon = \lambda_2/\lambda_1 - 1$ and $\tan \varphi_0 = \mathbf{v}_1^2/\mathbf{v}_1^1$ (or, equivalently, $\tan \varphi_0 = -\mathbf{v}_2^1/\mathbf{v}_2^2$), where $\lambda_{1,2}$ ($\lambda_1 < \lambda_2$) and $\mathbf{v}_{1,2}$ are, respectively, the eigenvalues and the eigenvectors of the tensor $\mathcal{I}_\varphi$. The original two-dimensional XRPD image together with the inertia principal axes are shown in Fig. 1(d).

2.3. Conformal mapping and integration

Once the relevant parameters ε and φ_0 have been obtained, the local isomorphism between $\mathbb{R}^2$ and the complex plane $\mathbb{C}$ can be expressed in polar (instead of elliptical) coordinates, *i.e.* $x + iy = \rho \exp(i\varphi)$, where ρ , defined as $a/[1 + \varepsilon \sin(\varphi - \varphi_0)]^{1/2}$, is known at this stage. After the determination of the detector tilt ε and rotation φ_0 , the original XRPD pattern is remapped onto a spherically symmetric pattern and the azimuthal invariance of the beam-sample-detector system is finally restored. Such a conclusion is crucial for the analysis.

We do not apply data-reduction procedures (detector response, diffraction corrections, Bragg peak removal *etc.*) at this stage; their inclusion will not affect our conclusions (see *e.g.* Hammersley *et al.*, 1996).

After the beam-sample-detector orthogonality has been restored, only counting statistics can violate the azimuthal invariance of the remapped two-dimensional XRPD pattern; thus $\Phi(x, y) \rightarrow \Phi[\log(z)]$, where z denotes $x + iy$, and the final radial (azimuthal) XRPD intensity profile follows the integration over the conformally remapped domains (see Figs. 2b and 2c). For the positioning of the radial integrated

Figure 2
Remapping of the XRPD pattern. (a) Original XRPD image. (b) Original annular domains. (c) Conformally remapped domains by means of a log function. (d) Conformally remapped XRPD image.

Figure 3

Radial/azimuthal intensity profiles. (a) Conformally remapped XRPD image. (b) Radial intensity profile (arbitrary units). (c) Comparison of XRPD radial intensity profiles corresponding to Fig. 2(a) obtained by our procedure and by the *Fit2D* software. The difference profile is plotted at the bottom. Units are arbitrary. (d) Azimuthal intensity profile (arbitrary units).

intensity profile on the resolution axis it is necessary to refine the detector-to-sample distance, which was fixed at the outset (see Cervellino *et al.*, 2006). The resulting intensity and azimuthal profiles corresponding to Fig. 2(d) are shown in Figs. 3(b) and 3(d), together with the corresponding one-dimensional intensity pattern extracted by the program *Fit2D* (Fig. 3c). The difference profile is also plotted (Fig. 3c), showing the good agreement between the patterns.

3. Results and conclusions

We have considered a new approach to the unfolding of two-dimensional images obtained from powder diffraction experiments using two-dimensional detectors. In this approach, the sample-to-detector distance is the only parameter that must be determined using a standard sample. Our method relies on the decoupling of two main problems: the beam-centre positioning and the calculation of eccentricity and rotation of the set of ellipses recorded. The two tasks are, respectively, accomplished with a self-convolution and an inertia tensor diagonalization. For the test sample the values for the eccentricity ε and the rotation φ_0 are, respectively, $0.87(11) \times 10^{-3}$ and $80(3)^\circ$, in good agreement with the average estimated values $0.93(10) \times 10^{-3}$ and $82(5)^\circ$ obtained by a completely different approach, a combination of simu-

lated annealing with Hankel–Lanczos singular value decomposition (Cervellino *et al.*, 2006); the present approach is completely analytical and much faster than the latter. A conformal mapping of the annular domains onto the radius–azimuth plane finally provides both the 2θ intensity profile and the azimuthal profile. This approach has been successfully applied on a set of XRPD CCD images of samples of the NIST standard Si 640c (Cline *et al.*, 2000) collected with a Nonius KappaCCD diffractometer.

References

- Cedola, A., Mastrogiacono, M., Burghammer, M., Komlev, V., Giannini, P., Cancedda, R., Rustichelli, F., Favia, A. & Lagomarsino, S. (2006). *Phys. Med. Biol.* **51**, N109–N116.
- Cedola, A., Mastrogiacono, M., Lagomarsino, S., Cancedda, R., Giannini, C., Guagliardi, A., Ladisa, M., Burghammer, M., Rustichelli, F. & Komlev, V. (2007). *Spectrochim. Acta B*, **62**, 642–647.
- Cervellino, A., Giannini, C., Guagliardi, A. & Ladisa, M. (2006). *J. Appl. Cryst.* **39**, 745–748.
- Cline, J. P., Deslattes, R. D., Staudenmann, J.-L., Kessler, E. G., Hudson, L. T., Henins, A. & Cheary, R. W. (2000). National Institute of Standards and Technology Standard Reference Material, https://srms.nist.gov/view_cert.cfm?srm=640C.
- Guagliardi, A., Giannini, C., Ladisa, M., Lamura, A., Laudadio, T., Cedola, A., Lagomarsino, S. & Cancedda, R. (2007). *J. Appl. Cryst.* **40**, 865–873.
- Hammersley, A. P., Brown, K., Burmeister, W., Claustre, L., Gonzalez, A., McSweeney, S., Mitchell, E., Moy, J.-P., Svensson, S. O. & Thompson, A. W. (1997). *J. Synchrotron Rad.* **4**, 67–77.
- Hammersley, A. P., Svensson, S. O., Hanfland, M., Fitch, A. N. & Häusermann, D. (1996). *High Pressure Res.* **14**, 235–248.
- Hanley, Q. S. & Denton, M. B. (2005). *J. Synchrotron Rad.* **12**, 618–625.
- Hinrichsen, B., Dinnebier, R. E. & Jansen, M. (2007). *Z. Kristallogr. Suppl.* **26**, 215–220.
- Mastrogiacono, M., Papadimitropoulos, A., Cedola, A., Peyrin, F., Giannini, P., Pearce, S. G., Alini, M., Giannini, C., Guagliardi, A. & Cancedda, R. (2007). *Biomaterials*, **28**, 1376–1384.
- Muchmore, S. W. (1999). *Acta Cryst. D* **55**, 1669–1671.
- Paris, O. & Riekel, C. (2005). *J. Synchrotron Rad.* **12**, 712.
- Phillips, W. C., Stewart, A., Stanton, M., Naday, I. & Ingersoll, C. (2002). *J. Synchrotron Rad.* **9**, 36–43.
- Rajiv, P., Hinrichsen, B., Dinnebier, R., Jansen, M. & Joswig, M. (2007). *Powder Diff.* **22**, 3–19.
- Rodriguez-Navarro, A. B. (2006). *J. Appl. Cryst.* **39**, 905–909.
- Suzuki, M., Yamamoto, M., Kumasaka, T., Sato, K., Toyokawa, H., Aries, I. F., Jerram, P. A., Gullick, D. & Ueki, T. (1999). *J. Synchrotron Rad.* **6**, 6–18.
- Takeda, A., Uekusa, H., Kubo, H., Miuchi, K., Nagayoshi, T., Ohashi, Y., Okada, Y., Orito, R., Takada, A. & Tanimori, T. (2005). *J. Synchrotron Rad.* **12**, 820–825.