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System of methods and programs for the characterization of axial textures by synchrotron light and electrons

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Crystallographic algorithms and computer programs play significant roles in materials' characterization. A software package for the quantitative characterization of crystallographic texture, under axial symmetry conditions, is presented. The proposed methodology is intended for use with both electrons and high-energy synchrotron X-rays. Three different programs are introduced. Anaelu and Grazing are based on the Rietveld modelling approach. Dianne follows the Bunge's symmetrized spherical harmonics method. A $SrTiO_3$ model sample is proposed as a hypothetical study case.

Keywords: Fibre texture; synchrotron; crystallographic software; pole figure; inverse pole figure.

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1. Introduction

Axially textured materials are frequently found in bulk and nanostructured functional materials. These types of samples are often characterized on electron microscopes, laboratory x-ray diffractometers and synchrotrons. The input data are the observed intensities of the diffraction peaks, either in a one-dimensional measurement (symmetric Bragg-Brentano θ - 2θ or grazing-incidence geometry) or in a two-dimensional detection experiment (transmission or grazing-incidence). The sought goal is the determination of the inverse pole figure (IPF), R(**h**), of the sample symmetry axis y_0 (h is a variable reciprocal vector). This work considers two paths to reach the proposed objective:

- The Rietveld approach [1]: The researcher proposes an initial combination of experimental parameters, crystal structure and inverse pole figure. Then he (she) models, with the help of the computer, the direct pole figures and the (one- or two-dimensional) XRD pattern and performs a fitting process leading to a refined description of the investigated polycrystal.

- The Bunge method [2]: The user converts a 2DXRD pattern into a set of direct pole figures (PFs), represents the pole figures as Legendre polynomials' expansions, calculates the inverse pole figure spherical harmonics expansion coefficients and synthesizes the desired IPF.

The work that has been done in relation to the Rietveld approach consists of the integration as a software package of the *Anaelu* [3] and *Grazing* [4] codes. The contribution to the Bunge methodology resides in the creation of the new *Dianne* program to invert direct pole figures by means of symmetrized spherical harmonics expansions. The developed codes may be downloaded from the Gitlab repository: `https://gitlab.com/cimav-tools`.

2. Fundamentals, methods and programs

2.1. Texture analysis, the Rietveld approach

2.1.1. 1D diffraction, program Grazing

The intensity of a diffraction maximum, for a sample with fiber texture, is given by Eq. (1):

$$I_h^{\text{Textured}} = I^{\text{random}} \cdot T. \tag{1}$$

The texture descriptor factor T, depending on the experimental configuration, is given by Eq. (2):

$$T = \begin{Bmatrix} P_h\theta_{\text{bragg}} & \text{Grazing incindence} \\ R(\mathbf{h}) & \text{Bragg-Brentano} \end{Bmatrix}, \tag{2}$$

where P_h (θ_{Bragg}) is the direct pole figure of the **h** = (h,k,l) planes, evaluated at the Bragg angle θ_{Bragg}. R(**h**) is the inverse pole figure function of the sample's axial symmetry axis. In the considered methodology, the proposition of a model inverse pole figure is required. It is generated by using a Gaussian distribution, Eq. (3):

$$R(\mathbf{h}) = R_0 e^{-4\log 2(\phi/\text{FWHM})^2}, \tag{3}$$

where ϕ is the angular distance between the reciprocal direction **h** and the Gaussian maximum, FWHM is the full width at half maximum of the orientation distribution. The maximum value of the distribution, R_0, is determined by a normalization condition. In fiber textures, the direct pole figures are obtained from the inverse one by applying the fundamental Equation of fiber textures, Eq. (4),

$$P_h(\phi) = \frac{1}{2\pi}\int_0^{2\pi} R(\phi,\psi)d\psi. \tag{4}$$

The presented mathematics have been systematized in the Grazing program. This program covers grazing incidence and Bragg-Brentano cases.

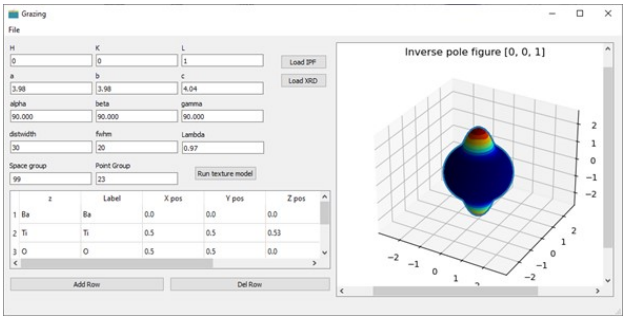

FIGURE 1. *Grazing* interface. The GUI displays the main controller of the software. As an example, the IPF of a hypothetical $BaTiO_3$ sample with $\mathbf{h}_0 = [0, 0, 1]$, FWHM $= 20^\circ$ is shown.

Texture model is based on Eq. (3). The modelled diffractograms are based on the theoretical intensities obtained with Eq. (1). Input data consists of crystal structure, preferred orientation vector $\mathbf{h}_0 = [h_0, k_0, l_0]$ and IPF full width at half maximum (FWHM). Based on mentioned data, *Grazing* obtains intensities without textures and applies the correct modulations for each method. For Bragg-Brentano method, the inverse pole figure, Eq. (3), modulates the intensities of peaks. In grazing incidence, the intensities' modulation is produced by the direct pole figures, calculated by Eq. (4).

Grazing is a dual language software. It utilizes the Fortran CrysFML library as a primary source for powder difractogram obtention, and Pyside2 as his graphic user interface (GUI) framework. Figure 1 shows a representative screen shot from the *Grazing* GUI.

Anaelu is currently developed on Python and Fortran. It utilizes the CrysFML library, similar to *Grazing* [5]. The majority of *Anaelu* math is devised on Fortran. The current graphic interface is based on QT framework, particularly on the Pyside2 [6] library. Its current graphic interface, which is compatible with Linux and Windows operative systems. is shown in Fig. 2.

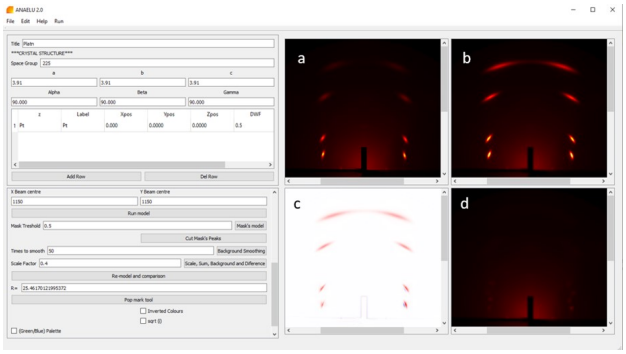

FIGURE 2. *Anaelu* interface. Graphic user interface (GUI) shows a complete virtual experiment of a Pt polycrystal with [1,1,1] sharp texture. The windows show: a) the experimental 2D XRD pattern, b) the modelled spectrum, c) an intermediate difference pattern and d) the subtracted background.

2.1.2. 2D diffraction, program Anaelu

Program *Anaelu* offers an approximation to a bidimensional Rietveld. It requires a model with an initial texture and leads, with an adjustment between calculated and experimental bidimensional diffractograms, into a complete characterization of the polycrystal considered.

For the interpretation of bidimensional diffractograms, *Anaelu* utilizes the 2D pattern to show the intensity distribution along the so-called Debye rings. The modulation of intensities is governed by the PFs, as shown in Eq. (2).

The calculated PFs are calculated from the proposed IPF with the fundamental equation of fiber textures, Eq. (2). Parameters' refinement is required for the quantitative 2D Rietveld.

2.2. Texture analysis, the Bunge approach. 2D diffraction, program *Dianne*

Bunge-based algorithm is used to calculate the inverse pole figure from direct figures obtained from the 2D Debye rings. The distribution of intensities along the ring is proportional to the corresponding pole figure $P_{(h_i)}(\phi)$. The mentioned condition is valid for small wavelengths, of the order of 0.02 Å or smaller. Mentioned wavelengths are obtained in high-energy synchrotrons and in transmission electron microscopes.

Program *Dianne* uses the properties of functional bases to decompose and invert a collection of PFs. PFs are represented as Legendre polynomials expansions and IPF as spherical harmonics expansions.

Using (5) we calculate the Legendre polynomials $\bar{\mathrm{P}}_1(\phi)$ expansion coefficients (F_1) of the direct pole figure $P_{(h_i)}(\phi)$. The inversion of the pole figures is via the equations' system (6). The determination of the inverse pole figure coefficients, C_l^m, allows the spherical harmonics $Y_l^m(h)$ synthesis of the inverse pole figure by means of Eq. (7).

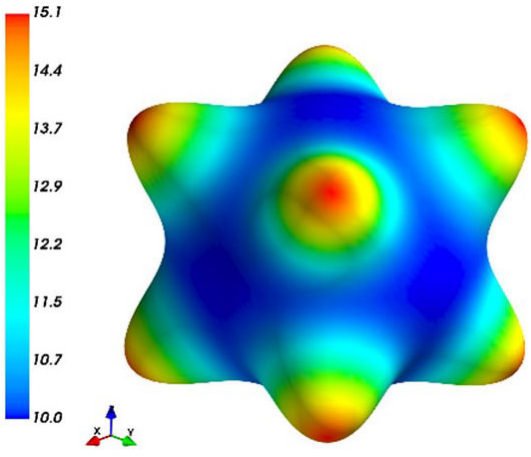

FIGURE 3. Inverse pole figure for a model axial texture. $SrTiO_3$, crystal point group m3m, IPF maxima at $< 1, 1, 1 >$, FWHM = 30°. Reference sphere radius = 10.

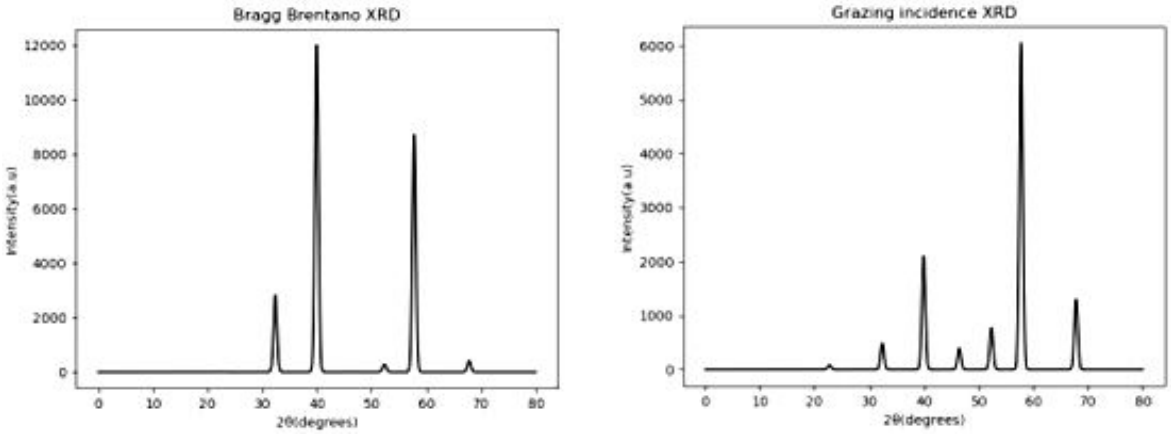

FIGURE 4. Grazing incidence vs Bragg Brentano in Grazing. Simulated 1DXRD patterns for a SrTiO3 virtual sample with $[1,1,1]$ texture, FWHM = 30°. a) Bragg-Brentano geometry. b) Grazing incidence diffractogram.

$$F_l(h_i) = \int_0^{\pi} P_{h_i}(\phi)\bar{P}_1(\phi)sin(\phi)d\phi, \tag{5}$$

$$F_l(h_i) = \sqrt{\frac{2}{2l+1}} \sum_{m=-l}^{l} C_l^m Y_l^m(h_i), \tag{6}$$

$$R(\phi,\psi) = \sum_{l=0}^{L_{\max}} \sum_{m=-1}^{l} C_l^m Y_l^m(\phi,\psi). \tag{7}$$

The determination of the inverse figures becomes more accurate by increasing the degree $L_{\max}$ of the spherical harmonics expansion. Same goes for the number of direct figures necessary to work as a function of $L_{\max}$. Symmetrized spherical harmonics [7], significantly reduce the number of required direct pole figures, by taking advantage of crystal symmetry.

Dianne is based on Python. It calculates the Legendre polynomials and the spherical harmonics by using the Scipy library [7].

3. Results and discussion

To exemplify the functionality of the created software package, the characterization of a hypothetical textured sample of $SrTiO_3$ is presented. The *in-silico* experiment recreates the investigation reported by Duran [8]. The material is interesting due to its electric properties. The assumed preferred orientation is [1,1,1] with FWHM = 30°.

3.1. Case of study: $SrTiO_3$ via *Grazing*

Equation (4) is used to calculate all the PFs and to evaluate them on the Bragg condition. *Grazing* multiplies the intensities of the untextured modelled diffractograms by the proper modulation factors (Eq. (2)) and calculates the 1DXRD patterns corresponding to Bragg-Brentano and grazing incidence configurations. Figure 4 shows the calculated XRD for both cases. In A) the intensities of the XRD peaks are multiplied by the inverse pole figure proposed by the user, evaluated in the reciprocal direction h_i=(θ,ϕ). In B) the intensities are multiplied by the direct pole figures, evaluated in the Bragg angle.

3.2. Case of study: $SrTiO_3$ via *Anaelu*

Given the crystal structure and the proposed texture of the considered perovskite, *Anaelu* models the theoretical 2D diffractogram. The assumed experimental configuration corresponds to a grazing incidence measurement, thus only the upper semi-circle of the 2DXRD pattern is observable. Figure 5 shows the calculated distribution of intensities along the Debye rings, as derived from Eq. (2). Fitting the calculated 2DXRD to the observed one allows the user to refine the crystal structure-texture configuration.

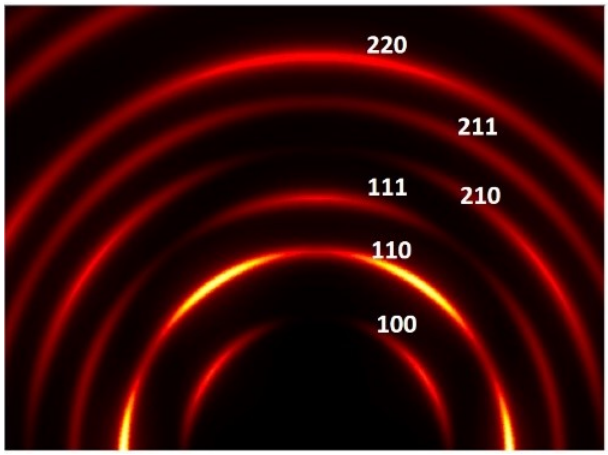

FIGURE 5. Simulated 2D XRD for textured $SrTiO_3$.Patter modelled by *Anaelu*.

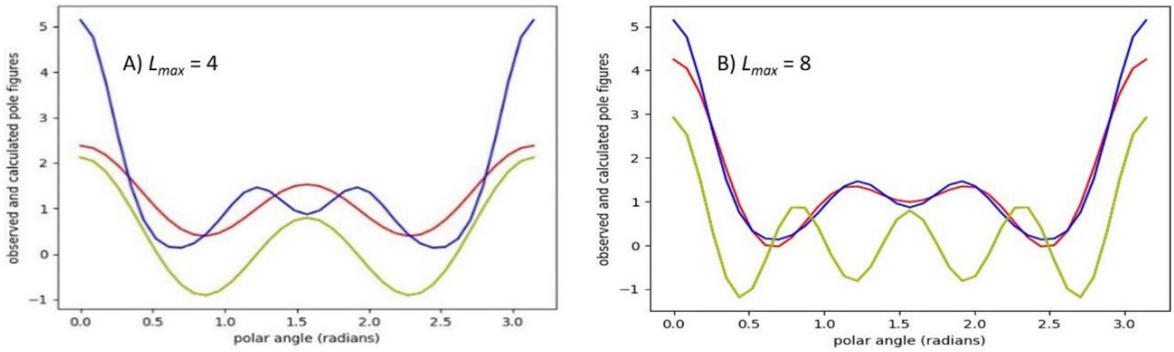

FIGURE 6. Legendre approximation to $SrTiO_3$ $[1,1,1]$ pole figure. Colors code: blue = "observed" PF; green = Legendre polynomial for $l = L_{\max}$; wine = expanded representation.

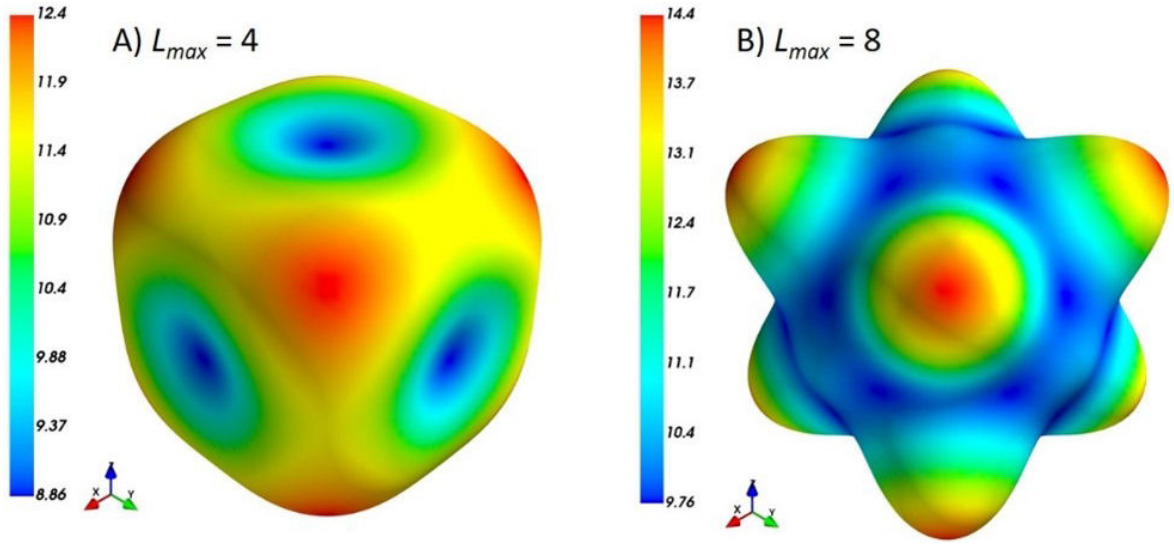

FIGURE 7. Symmetrized spherical harmonics expansion of $SrTiO_3$ IPF.

3.3. Case of study: SrTiO3 via *Dianne*

Since the material possess a cubic symmetry, *Dianne* is well fitted for application of Bunge's symmetrized spherical harmonics [2]. The considered PFs were [1,0,0], [1,1,0] and [1,1,1]. Figure 6 exemplifies the representation of the [1,1,1] PF as a Legendre polynomial expansion. In A) the expansion up to $\mathbf{L_{max} = 4}$ is shown. B) displays the improvement of the description for $\mathbf{L_{max} = 8}$.

The same process was applied to PFs [1,0,0] and [1,1,0]. The expansion coefficients of the mentioned PFs, $F_l(h_i)$, were used to build the equations system (6). Solving these equations leads to calculate the IPF expansion coefficients C_l^m. Figure 7 shows the progressive synthesis of the $SrTiO_3$ IPF by the mentioned procedure.

4. Conclusions

The presented software package allows two paths for the characterization of axial textures, starting from 1D or 2D diffraction experiments. The *Grazing* - *Anaelu* programs apply 1D and 2D Rietveld approaches. These consist on proposing an initial combination of crystal structure and texture, followed by a IPF → PFs → XRD calculation and refinement. *Dianne* systematizes the Bunge methodology: Experimentally measured pole figures are inverted via a symmetrized spherical harmonics algorithm. The application of the proposed software package to an illustrative case of study has been exposed. The consistency of the programs' system has been shown.

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