MATHEMATICAL PROPERTIES OF DIFFRACTION POLE FIGURES

Helmut Schaeben

Mathematics and Computer Sciences in Geology,
Department of Geology, Freiberg University of Mining and Technology,
Freiberg, Saxony, FRG

ABSTRACT

Crystallographic diffraction pole figures are defined as hyperspherical x-ray transforms of an orientation density function. The range of this transform is characterized in terms of an ultrahyperbolic differential equation which is derived without referring to spherical harmonics. The general solution of the differential equation is given both in terms of its characteristics or spherical harmonics.

These results will aid in the solution of actual problems of texture goniometry, e.g. the high-resolution analysis of preferred crystallographic orientation in polycrystalline materials by diffraction; it is shown in particular that solving the inverse tomographic problem of texture goniometry and determining additional pole figures which have not been measured are two equivalent issues of the same problem. Eventually, the paradigm [1] that the determination of additional pole figures requires first the determination of an reasonable orientation density function is shown to be obsolete.

POLE FIGURES, HYPERSPHERICAL X-RAY TRANSFORMS

The analysis of preferred crystallographic orientation is referred to as texture analysis. A probability density function \( f(g) \) defined on the group \( SO(3) \) of rotations or a subset \( G \) of \( SO(3) \) depending on the crystal symmetry, respectively, is referred to as orientation density function describing the distribution of crystallographic orientations by volume. While orientation density functions may be mathematically determined from individual orientation measurements directly in some applications, they are not generally directly accessible in many other applications. In these applications it is common practice to measure diffraction pole figures of a few crystallographic forms \( h \).

A diffraction pole figure is mathematically represented as the projection of an orientation density function basically provided by the integral operator

\[
(\mathcal{P}_h f)(r) = \frac{1}{2\pi} \int_{\{g \in SO(3)\} | \mathbf{h} = \mathbf{g}r\}} f(g) \, dv(g) = P(h, r)
\]

where the function \( P(h, r) \) for a given crystallographic direction \( h \in S^2 \) may be referred to as hyperspherical x-ray transform of \( f \) with respect to \( h \). The path of integration \( \{g \in SO(3) \} \)
This document was presented at the Denver X-ray Conference (DXC) on Applications of X-ray Analysis.

Sponsored by the International Centre for Diffraction Data (ICDD).

This document is provided by ICDD in cooperation with the authors and presenters of the DXC for the express purpose of educating the scientific community.

All copyrights for the document are retained by ICDD.

Usage is restricted for the purposes of education and scientific research.

DXC Website — www.dxcicdd.com

ICDD Website - www.icdd.com
In eq. (1) is a circle restricted to the three-dimensional sphere $S^3$ in four-dimensional Euclidean space $\mathbb{R}^4$ [10].

Next, for reasons originating in crystallography and Friedel's empirical law, which states that the diffracting x-ray cannot distinguish between top and bottom of the lattice planes, i.e. the directions $h$ and $-h$ cannot be distinguished in the diffraction experiment even if the crystal itself is not centrosymmetric, the basic crystallographic x-ray transform is defined as

$$\left(\tilde{\mathcal{P}}_h f\right)(r) = \frac{1}{2} (P(h, r) + P(-h, r)) = \tilde{P}(h, r)$$

where $S^2_+$ denotes the upper unit hemisphere in $\mathbb{R}^3$.

Now, obviously,

$$\tilde{P}(h, r) = \tilde{P}(-h, r) = \tilde{P}(h,-r)$$

that is, $\tilde{P}$ is an even function in both arguments $h, r \in S^2$, and therefore the operator of eq. (2) maps essentially from $\text{SO}(3)$ (equivalent to $S^4_+$) onto the cross-product $S^2_+ \times S^2_+$ of two upper hemispheres, or equivalently from the projective space $\mathbb{H}^3$ to the cross-product space $\mathbb{H}^2 \times \mathbb{H}^2$ of projective planes.

Let $G_B = \mathcal{G}_B \subset \text{SO}(3)$ denote the finite point subgroup of proper rotations associated with the crystal symmetry class $\mathcal{G}_B$. Due to Friedel's law the effective crystal symmetry is described by the point group $\mathcal{G}_B = \mathcal{G}_B \otimes \{1,-1\}$, where 1 denotes identity and -1 the symmetry operation of inversion, which is also referred to as Laue class. Corresponding to the 32 crystal classes there exist 11 Laue classes $\mathcal{G}_B$, which always contain the operation of inversion as an element of symmetry.

The 32 different symmetry classes $\mathcal{G}_B$ are given by 11 purely rotational groups and 11 Laue groups; for both $\mathcal{G}_n = G_B \otimes \{1,-1\}$, thus $\mathcal{G}_n = G_B$ holds. For the remaining 10 groups $\mathcal{G}_n \neq G_B \otimes \{1,-1\}$ and hence $\mathcal{G}_n = \mathcal{G}_B \subset \text{SO}(3) = (G_B \otimes \{1,-1\}) \subset \text{SO}(3) \neq G_B$; considering only $G_B$ instead of $\mathcal{G}_B$ means an essential loss of information in these cases, cf. [7].

Let $g$ be an arbitrary element of $\text{SO}(3)$. In case of crystallographic symmetry the (right) coset of orientations $G_B g = \{ gBg \mid gB \in G_B \}$, consists of orientations which are physically indistinguishable with respect to the coordinate system $K_A$ fixed to the sample; $g$ is referred to as a representative of the coset $G_B g$. The cosets with respect to $G_B$ provide a partition of $\text{SO}(3)$. Therefore, the property
\(f(g_B g) = f(g)\) for all \(g_B \in G_B\) \hspace{1cm} (4)

is imposed on \(f\), that is, \(f\) is constant on each coset. Eventually, it will be sufficient to consider \(f\) on an elementary set \(G\) of representatives of each coset, i.e. on the set of physically distinct orientations.

For a function \(f\) with the property (4) their corresponding x-ray transforms satisfy

\[P(g_B h, r) = P(h, r)\] \hspace{1cm} (5)

For an orientation density function \(f \in E^2(\text{SO}(3))\), its corresponding crystallographic \(h\) pole figure density function \(\tilde{P}_h\) of the crystal form \(h = \{h_m | m=1, \ldots, M_h\} \subset S^2\) of multiplicity \(M_h\) corresponding to crystal-symmetrically equivalent lattice planes \(\{(hk\ell)_m | m=1, \ldots, M_h\} \subset \mathbb{Z}^3\) is defined for \(r \in S^2\) as

\[
\tilde{P}_h(r) = (\tilde{\alpha}_h f)(r) - \frac{1}{M_h} \sum_{m=1}^{M_h} \tilde{P}(h_m, r)
\] \hspace{1cm} (6)

Summation in (6) is effectively over all directions \(g_B h, -g_B h, g_B \in G_B\), which are symmetrically equivalent with respect to the associated Laue group \(g_B\). Eventually, applying eqs (4) and (5)

\[
(\tilde{\alpha}_h f)(r) = \frac{1}{\# G_B} \sum_{g_B \in G_B} \tilde{P}(g_B h, r) = \tilde{P}(h, r)
\] \hspace{1cm} (7)

where \(h\) denotes the set of symmetrically equivalent crystallographic directions \(h_m\) related to each other by the associated Laue group, and \(h\) denotes an arbitrary element of \(h\). Thus, \(\tilde{P}_h(r)\) and \(\tilde{P}(h, r)\) will no longer be distinguished.

In the past, main interest of texture analysis was focused on the inverse problem to find a reasonable orientation density function given intensity data corresponding to pole figures of some crystal forms. It was not appreciated that pole figures are rather complex mathematical entities governed by a differential equation of ultrahyperbolic type.

THE DIFFERENTIAL EQUATION GOVERNING POLE FIGURES
Let $A$ denote the Laplace-Beltrami operator, and assume $f$ and hence $P$ is twice continuously differentiable. Then

$$\left(\Delta_h - \Delta_r\right)P(h,r) = 0$$  \hspace{1cm} (8)

where the equality is directly derived from the definition, eq. 1. It is referred to as ultrahyperbolic differential equation in mathematical physics [3], [6], or Darboux partial differential equation in tomography [4]. In texture goniometry it was first noticed in [9].

In complete analogy to (i) Fourier's harmonic method and (ii) d'Alembert's method of characteristics to solve an ordinary hyperbolic differential equation, e.g. to solve the Cauchy problem of mathematical physics [2], the general solution of the ultrahyperbolic differential equation for hyperspherical x-ray transforms is obtained both in terms of (i) spherical harmonics and (ii) its characteristics.

The general solution of eq. (8) in terms of harmonics is

$$P(h,r) = \sum_{l=0}^\infty \sum_{m=-l}^l \sum_{n'=l}^l C_{l,m}^{m'}(h)Y_{l,m}(r)$$  \hspace{1cm} (9)

Physical requirements like non-negativity, evenness, crystallographic or sample symmetries impose additional constraints on the system eq. (9) and lead to special solutions. It is emphasized that eq. (9) relates pole figures directly to $C$-coefficients, implying that $C$-coefficients and thus additional pole figures can directly be determined from pole figure data.

Conversely, any function satisfying a differential equation (8) is shown to be the x-ray transform of a function under mild mathematical assumptions. More specifically, for any function $u \in \mathcal{C}^\infty(S^2 \times S^2)$ satisfying the differential equation (8), there exists a unique function $f \in \mathcal{C}^\infty(SO(3))$ such that $\mathcal{P}_h f = u$.

It should be noted that if $u \in \mathcal{C}^\infty(S^2 \times S^2)$ satisfying eq. (8) is even in both arguments, then a unique even function $f \in \mathcal{C}^\infty(SO(3))$ exists such that $\mathcal{P}_h f = u$. Since the crystallographic x-ray transform $\mathcal{P}_h f$ of $f$ is always even, only the even part of $f$ is uniquely determined.

Thus, the coefficients of the general solution of the ultrahyperbolic equation for spherical x-ray transforms in terms of harmonics are the harmonic coefficients of the function being transformed, i.e. the solution of the ultrahyperbolic equation provides the solution of the inverse x-ray transform problem. Whenever it is possible to determine the even part $\tilde{f}$ of an orientation density function conforming with given crystallographic pole figures of some crystal forms, then it is possible to determine directly the pole figure of any other crystal form, and vice versa. This possibility to solve a differential equation instead of an inverse
Radon-type problem has already been shown elsewhere. In particular, this duality may be exploited to improve on the conventionally obtained harmonic solution of the inverse problem of texture analysis.

Any function \( u(h, r) \in C^2(S^2 \times S^2) \) which depends on the dot product \( h \cdot r = \cos \eta \) only obviously satisfies the differential equation (8). Furthermore, an appropriate linear transformation of \( h \) or \( r \), respectively, with constant coefficients should not change this situation. Consequently, the characteristics of the ultrahyperbolic differential equation are the fibres \( h \cdot g r = \text{const} \), where \( g \) is an arbitrary proper rotation.

Thus, the general solution of equation (8) in terms of characteristics can be represented as

\[
u(h, r) = \sum_l \sum_k u_l(h \cdot g_k r)\tag{10}
\]

where \( g_k \in SO(3) \) are arbitrary rotations and \( u_l \in C^2(\mathbb{R}^l) \) are some real twice continuously differentiable functions. A specific solution is constructed analogously to d'Alembert's method of characteristics by choosing distinguished functions \( P_l \) for the functions \( u_l \) and fitting them to the initially given pole density figures, where the \( P_l \) belong to some specific dense subset of \( C^2(\mathbb{R}^l) \), e.g. polynomials, splines, wavelets or radial basis functions.

In a particularly simple case, a twice continuously differentiable function \( u \) satisfying eq. (8) is considered, which reduces to a function of \( h \cdot g_0 r \) for some arbitrary fixed \( g_0 \in SO(3) \). Then \( u \) is uniquely determined provided for some given \( h_0 \in S^2 \) it is known for all \( g_0 r \) on a great circle containing \( h_0 \).

With respect to practical texture analysis these assumptions may appear rather artificial; nevertheless, this proposition is the basis of texture component fit methods [5], [10]. Moreover, the approach by characteristics can be greatly generalized.

It can generally be shown that a series expansion into appropriately chosen functions \( p_l \) is equivalent to the series expansion into spherical harmonics. Since any continuously differentiable function on the sphere \( S^2 \) can be expanded into an absolutely convergent series of spherical harmonics, there exists an equivalent expansion into linear combinations of specified spherical harmonics, the argument of which is the scalar product of \( h \) and \( r \). This expansion provides the means to represent any pole density function in the form (10).

For a more complete discussion of the subject including proofs the reader is referred to [8].

CONCLUSIONS

It is shown that the approaches to texture analysis provided by the projection formula or the differential equation are equivalent, but put emphasis on different issues of the same problem. Eventually, it is concluded that whenever it is possible to calculate a reasonable
approximate of the even portion of an orientation density function conforming with experimental diffraction pole figures of given crystal forms, then it is possible to determine reasonable approximates of the pole figures of any other crystal form directly from the given ones, and vice versa.

Moreover, this equivalence provides the link to mathematical tomography and suggests to refer to texture analysis as an application of spherical mathematical tomography. Pursuing this approach should result in the design of improved measuring strategies and improved methods analysis and interpretation.

REFERENCES