Diffraction tomography inversion and the transverse ray transform

Lionheart, William R.B. and Korsunsky, Alexander M.

2021

MIMS EPrint: **2021.11**

ISSN 1749-9097

Diffraction tomography inversion and the transverse ray transform

William R.B. Lionheart[1] and Alexander M. Korsunsky [2]

[1]Department of Mathematics, University of Manchester, UK
[2]Department of Engineering Science, University of Oxford, UK

Abstract We show that a reciprocal space squared intensity map of a material can be recovered, for each characteristic length scale, from diffraction tomography data by a simple slice-by-slice reconstruction method. Moreover if the reciprocal space map can be represented by a finite sum of spherical harmonic components for each length scale then the coefficients of that expansion can be recovered from inverting the transverse ray transform (TRT), where the data are polynomial coefficients of the azimuthal diffraction pattern for each length scale.

1 Introduction

X-ray diffraction experiments give information about the structure of a material on the length scale of the wavelength X-rays used. In X-ray crystallography a periodic crystal structure gives rise to a periodic diffraction pattern with distinct peaks. For less regular materials a less distinct diffraction pattern can never-the-less detect preferred orientations and nearly periodic structures.

If a narrow gauge volume is illuminated with a monochromatic X-ray beam the diffraction pattern is a sum of diffraction patterns in that volume [1]. Increasingly, not just for X-rays but also neutrons and electrons, we have the capability to raster scan a narrow beam measuring a diffraction pattern and perform a combination of tomography and diffraction, hoping to reconstruct a 3D diffraction pattern that that summarizes the properties of the material in each voxel. Small angle X-ray scattering (SAXS) tomography is a particularly promising variant of this idea. However so far there is no theory for the reconstruction in this field, and although it has been assumed so, it is not yet proven that an isotropic average can be meaningful reconstructed from this data. In this paper we lay the theoretical foundation for diffraction tomography. We will demonstrate a theoretical reconstruction method using data from all directions. We will also show how a slice-by-slice approach can be used to reconstruct a diffraction pattern given by a finite sum of spherical harmonics. In this case the problem reduces to the transverse ray transform of symmetric tensor fields.

2 Physical model

We assume that at each point $\mathbf{x}$ in the object and for each three dimensional reciprocal space vector $\mathbf{q}$ there is a scattering intensity-squared map $f(\mathbf{x},\mathbf{q})$. For a given ray direction $\xi \in S^2$ (the unit sphere) a diffraction pattern for the material near $\mathbf{x}$ would produce a 2D scattering pattern on a planar detector normal to ξ with squared intensity $f(\mathbf{x},\mathbf{q})$ for $\mathbf{q} \in \xi^\perp$ (the space of vectors perpendicular to ξ). Clearly there is an underlying assumption that the problem can be formulated on two length scales, and we will not make this explicit mathematically, but roughly we are assuming that on the scale of the wave length of the X-rays (or particles) the 3D distribution of scatterers has $f(\mathbf{x},\mathbf{q})$ as the square magnitude of its Fourier transform and this can be treated as a constant on a small length scale, but on a larger length scale, commensurate with the width of the beam and the spatial scanning increments, the Fourier transform is variable.

We treat intensity squared as the variable as we assume that the diffraction pattern observed from one ray is an incoherent average and so the result of the sum of squared intensities along the gauge volume. Note that as f is magnitude squared Fourier transform of a real function it is even with respect to $\mathbf{q}$: $f(\mathbf{x},\mathbf{q}) = f(\mathbf{x},-\mathbf{q})$.

Our data then is the generalized transverse ray transform (GTRT)

$$g(\mathbf{x},\xi,\mathbf{q}) = \int_{-\infty}^{\infty} f(\mathbf{x}+s\xi,\mathbf{q})\,\mathrm{d}s, \tag{1}$$

for $\mathbf{x} \in \mathbb{R}^3, \xi \in S^2, \mathbf{q} \in \xi^\perp$. In the case where $f(\mathbf{x},\mathbf{q}) = F\cdot\mathbf{q}^m$ where F is a rank m symmetric tensor field (the dot denotes contraction over m indices) this coincides, after a small change in notation, with the transverse ray transform of symmetric tensor fields defined by Sharafutdinov [2].

3 Uniqueness and reconstruction for complete data

Suppose that we have diffraction data for all rays passing through the object (the support of f). For simplicity consider a single value of $|\mathbf{q}| = Q$, corresponding physically to one reciprocal length scale, and a circle on the detector plane of radius Q centred on its intersection with the ray. We now follow the same argument used by [2, p119] for the transverse ray transform of symmetric tensor fields. Choose a direction $\eta \in S^2$, which conceptually we think of as a rotation axis for the sample in an experiment. Now consider measurements of g for all rays in directions $\xi \in \eta^\perp$.

For a given plane through $\mathbf{x}_0+\eta^\perp$, through $\mathbf{x}_0$ normal to η, $g(\mathbf{x},\xi,Q\eta)$ for $\mathbf{x}\in\mathbf{x}_0+\eta^\perp, \xi\in\eta^\perp\cap S^2$ is the 2D X-ray transform of the scalar function $f(\mathbf{x},Q\eta)$ on that plane. Hence it can be reconstructed using the inverse Radon transform. We see now that f can be reconstructed from complete data g for all rays. In practice only reciprocal length scales $0<Q_0<Q<Q_1$ in some fixed range would make physical sense. We note that this idea is already present in the SAXS

tomography literature, for a single axis [3], and for multiple axes [4, 5] without formally describing the generalized transverse ray transform. In the mathematical literature the extension of ray transforms to the *sphere bundle* (space with a sphere at each point) appear as the geodesic ray transform on a Riemannian manifold.

4 Consistency conditions

In inverse problems in general and especially in tomography it is important to characterize data that is consistent with the assumed model: in mathematical terms, to describe the range of the operator. For some cases the singular value decomposition (SVD) gives an explicit orthogonal basis for the range and for its orthogonal complement. For the scalar x-ray transform in two dimensions see [6] and three dimensions see [7]. Consistency conditions are systems of equations that characterize the range. For the 2D Radon (X-ray) transform Helgason's range conditions characterize the range in terms of moments of the data [8, Thm 4.2]. For the 3D X-ray transform the range is characterized by satisfying John's ultrahyperbolic partial differential equation (PDE) [9].
For the isotropic case f independent of $\mathbf{q}$, the GTRT (1) reduces to the X-ray transform in 3D space. This is formally overdetermined as the space of lines in 3D space is four dimensional. The data is one function of four variables and we seek a function of three variables. So it is no surprise that the data satisfies one PDE. By contrast in the 2D isotropic problem (Radon transform) we seek one function of two variables and our data is one function of two variables — formally correctly determined. In the case of the GTRT we seek one function of five variables (for fixed Q). Our data is a function on a circle for each line, and is also a function of five variables, so formally correctly determined.
In Sec 3 we saw how one can reconstruct $f(\mathbf{x},\mathbf{q})$ on each plane normal to $\mathbf{q}$ as a scalar Radon transform on each plane. Notice for a fixed $\mathbf{q}$ the only data involving $f(\mathbf{x},\mathbf{q})$ is exactly the lines in the plane through $\mathbf{x}$ normal to $\mathbf{q}$. So Helgason's range conditions are the only consistency conditions that apply, beyond that g is even in $\mathbf{q}$. Data g satisfying these consistency conditions is associated with a (unique) f.

5 The Transverse ray transform of tensor fields

The transverse ray transform of a symmetric tensor field is the integral along rays of the projection of the ray normal to that direction. Let e_i be the unit Cartesian vectors in 3-space. We will denote the tensor product of tensors a and b by $a \otimes b$. A general rank two tensor has the form

$$a = \sum_{i,j=1,..,3} a_{ij} e_i \otimes e_j$$

We denote symmetric tensor product $a \odot b = (a \otimes b + b \otimes a)/2$ and the symmetric k-th tensor power by a^k. Let $\xi \in S^2$ be any unit vector, then the matrix

$$\Pi_\xi = I - \xi\xi^T$$

projects a vector on to the subspace $\xi^\perp$ or $(\Pi_\xi)_{ij} = \delta_{ij} - \xi_i \xi_j$ as a tensor. For a rank k symmetric tensor a the projection $P_\xi(a)$ is the k fold contraction of a with Π_ξ. In components

$$P_\xi(a)_{i_1 \cdots i_k} = \sum_{j_1 \cdots j_k} (\Pi_\xi)_{i_1 j_1} \cdots (\Pi_\xi)_{i_k j_k} a_{j_1 \cdots j_k}$$

for example as a matrix the components of the projection of a rank two symmetric tensor $P_{e_3} a$ are

$$\begin{bmatrix} a_{11} & a_{12} & 0 \\ a_{12} & a_{22} & 0 \\ 0 & 0 & \end{bmatrix}$$

For a rank k symmetric tensor field a the Transverse ray Transform (TRT) is defined as

$$Ja(\mathbf{x},\xi) = \int_{-\infty}^{\infty} P_\xi(a)(\mathbf{x} + s\xi)\,\mathrm{d}s,$$

note that the data for each ray defined by $\mathbf{x},\xi$ is a symmetric rank k tensor in three variables. However it is restricted to $\xi^\perp$ so it would be natural to express it in a two dimensional coordinate system for actual measurements (such as detector screen coordinates).
We can now review the known theory for sufficiency of data, characterization of consistent data and inversion for the TRT. Sharafutdinov [2, p119] (and earlier Russian edition) gives an inversion method for the TRT of a symmetric rank k tensor field that is equivalent to the argument we gave in Sec 2 applied to the special case

$$f(\mathbf{x},\mathbf{q}) = a(\mathbf{x}) \cdot \mathbf{q} \cdots \mathbf{q}$$

where the dots denote contraction and the result of the k fold contraction is a scalar.
As before we consider a rotation axis η and rays in directions $\xi \in \eta^\perp$, in each plane $\eta^\perp + z\eta$ the component $a(\mathbf{x}) \cdot \eta \cdots \eta$ (k-fold contraction) transforms as a scalar in the plane. We perform the reconstruction by application of the scalar inverse Radon transform to $Ja(x,\xi) \cdot \eta \cdots \eta$. One then has to repeat for at least $K = \binom{k+2}{2}$ (the dimension of the space of symmetric rank k tensors, sometimes called the 'stars and bars' problem) choices $\eta^1, .., \eta^k$, such that the set of symmetric k fold products $(\eta^i)^k$ is linearly independent. For example for $k = 2$ the six diagonals of the icosahedron is a suitable choice (in fact optimal as it maximizes the condition number of an associated linear system). See [10] for a geometric criterion for $k = 2$.
For modest k this still seems rather wasteful in that data is discarded and therefore more rotation axes are need than is strictly necessary. In [10] we gave a filtered back projection formula for the reconstruction of a rank-2 tensor from *complete* TRT data, that is rotation about *every* axis. This is even

more wasteful for small k however it does use all the data that can be collected, averaging over the redundancy.

In an attempt to reduce the number of rotation axes needed, in [11] we showed that there is an explicit reconstruction algorithm for the TRT for a rank-2 tensor using only three rotation axes. However this has a certain type of instability compared to using six axes.

6 Spherical harmonic expansion

It has been suggested (see eg [12],[13]) that the intensity squared reciprocal space map be expanded in spherical harmonics. Suppose we have

$$f(\mathbf{x},\mathbf{q}) = \sum_{l\le K, l\text{even}, |m|\le l} a(\mathbf{x},|\mathbf{q}|)_{lm} Y_l^m(\mathbf{q}/|\mathbf{q}|). \qquad (2)$$

Where we use the abbreviated notation for Laplace's spherical harmonics $Y_l^m(\hat{\mathbf{q}})$ for $Y_l^m(\theta,\phi)$ where $\hat{\mathbf{q}} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ is a unit vector. In [12] the complex reciprocal space map (not the square magnitude) is represented as a sum of spherical harmonics up to some order K. As the product of spherical harmonics can be expressed in spherical harmonics up to K we lose no generality.

The question arises if we can deduce the coefficients $a(\mathbf{x},Q)_{lm}$ from less than the full data $g(\cdot,\cdot,\mathbf{q})$ with $|\mathbf{q}| = Q$. In particular can the isotropic term a_{00} be deduced from averages over the circles of radius Q of the diffraction patterns? More generally can the components of each order be reconstructed separately by some form of preprocessing of diffraction pattern data? To answer these questions we need to consider the relationship between spherical harmonics and polynomials.

A homogeneous degree k polynomial on $\mathbb{R}^3$ is a polynomial p satisfying $p(c\mathbf{q}) = c^k p(\mathbf{q})$, In the discussion of polynomials we will use $\mathbf{q} = (q_1,q_2,q_3)$ as our general vector is in reciprocal space. For example $p(\mathbf{q}) = q_1^3 - 2q_2^2 q_3$ is a 3-rd degree homogeneous polynomial. Homogeneous polynomials of degree k are in one to one correspondence with symmetric k-th rank tensors, we just replace the q_i by unit basis vectors e_i and treat the product as the symmetric tensor product. For example $q_1^2 + q_2^2 + q_3^2$ corresponds to the Kroneker tensor with components δ_{ij}.

A harmonic polynomial is $p(\mathbf{q})$ is simply a polynomial satisfying Laplace's equation

$$\Delta_{\mathbf{q}} p(\mathbf{q}) = 0, \quad \Delta_{\mathbf{q}} = \frac{\partial^2}{\partial q_1^2} + \frac{\partial^2}{\partial q_2^2} + \frac{\partial^2}{\partial q_3^2},$$

for example $q_1^2 - q_3^2$ is harmonic.

The dimension of the space of spherical harmonics of degree l in 3 variables is $2l+1$ [14, prop 5.8)]. The Laplace spherical harmonics $Y_l^m(\mathbf{q})$ span the space of harmonics polynomials of degree l.

Our aim is to convert (2) to a tensor expression so that we can apply the known theory of tensor tomography. The problem is that while we can regard spherical harmonics as polynomials and polynomials as symmetric tensors we appear to have a sum of tensors of different ranks. To get around this first we impose the condition $|\mathbf{q}| = Q$, a reciprocal length scale. The expression

$$f_Q(\mathbf{x},\mathbf{q}) = \sum_{l\le K, l\text{even}, |m|\le l} a(\mathbf{x},Q)_{lm} |\mathbf{q}|^{K-l} Y_l^m(\mathbf{q}) \qquad (3)$$

is the a homogeneous polynomial of degree K in $\mathbf{q}$ at each $\mathbf{x}$. This polynomial has an associated rank K symmetric tensor field we will call $F_Q(\mathbf{x})$, and $f_Q(\mathbf{x},\mathbf{q}) = F_Q \cdot \mathbf{q}^K$.

Our task now is to show how the TRT data for F_Q can be recovered from g restricted to $|\mathbf{q}| = Q$. It is well known that a bilinear function $B(v,w)$ in two vector variables can be recovered from the quadratic form $P(v) = B(v,v)$ using the polarization identity

$$B(v,w) = \frac{1}{4}\left(P(v+w) - P(v-w)\right). \qquad (4)$$

It is perhaps not surprising, but less well known, that a similar identity applies to symmetric multi-linear functions [15]. The relevance to us is that for each ray and a given Q we know $g(\mathbf{x},\xi,\mathbf{q})$ for $|\mathbf{q}| = Q$, $\mathbf{q} \in \xi^\perp$, the diffraction pattern around a circle of radius Q. This is the integral of $F_Q \cdot q^m$ along a ray and we can find the TRT $JF_Q(\mathbf{x},\xi)$ by applying the multi-linear polarization identity to $g(\mathbf{x},\xi,\mathbf{q})$.

As long as it is known (2) is valid for some K one can attempt a reconstruction using the known reconstruction methods for the TRT detailed in Sec 5, or using regularized iterative methods widely used for large scale linear inverse problems: CGLS on an augmented matix for generalized Tikhonov and FISTA when a TV regularized term is included. These are implemented, for example, in our Core Imaging Library [16] for scalar problems. If K is not known *a priori* one can use a higher value than necessary, at the expense of higher computational cost, and then decide if the coefficients a_{lm} are significant if including them results in a significantly better fit to the data.

One tempting approach that may well fail is to take an average in the detector plane over a circle of constant Q and then attempt a slice by slice reconstruction assuming a scalar (that is isotropic) model with $K = 0$. The underlying problem is that the restriction of a harmonic polynomial in three variables to a plane is not necessarily harmonic. For example $q_1^2 - q^3$ is harmonic in three dimensional space but its restriction to $q_3 = 0$ is q_1^1 which is not harmonic. The projection on to spherical harmonic components of each order is not preserved by projection on to a plane.

In HAADF-STEM (High-Angle Annular Dark-Field Scanning Transmission Electron Microscope) tomography (see for example[17] [18]) an azimuthal average of a diffraction pattern is used to reconstruct a scalar image from a single tilt-series. Our analysis suggests this is flawed where the electron diffraction pattern is anisotropic.

To some extent the danger of assuming isotropy, or indeed too small a K in general, is reduced provided enough data is collected. For example if slice-by-slice data is collected for one rotation axis and a scalar reconstructed that best fits that data, one can then test if the same scalar reconstruction is consistent with reconstruction from rotation about a different axis.

On each plane normal to a vector η the contractions of Ja with η appear as the TRT of lower rank tensor fields on $\eta^\perp$. The range of the 2D TRT is given completely by the SVD described by [19].

7 Conclusions and further work

We have laid the theoretical framework for diffraction tomography including an explicit inversion procedure, for each reciprocal length scale, for *full data*, that is a sufficiently dense sampling of the four dimensional space of lines. We have also shown that assuming the reciprocal space map for each reciprocal length scale can be expanded in even spherical harmonics up to some fixed degree is equivalent to reconstructing a symmetric tensor field using the transverse ray transform data. The next steps practically are to do a full regularized algebraic reconstruction on real data, for both complete data, and with limited data assuming a finite spherical harmonic expansion. While an explicit reconstruction formula for limited TRT data is available for rank two tensors, none have been derived for higher rank tensors. Recent results on the TRT for higher rank tensors and limited data focus on the divergent beam case [20] and this application may provide the impetus needed for further work on the parallel beam case relevant to synchrotron X-ray (SAXS), electron (HAADF-STEM) and neutron (SANS) diffraction tomography[21].

Acknowledgements

WL would like to thank Marianne Liebi for helpful discussions, and to thank the Royal Society for a Wolfson Research Merit Award. Both authors gratefully acknowledge support from EPSRC grants EP/V007742/1 and EP/V007785/1 “Rich nonlinear tomography for advanced materials”.

References

[1] A. M. Korsunsky, N. Baimpas, X. Song, et al. “Strain tomography of polycrystalline zirconia dental prostheses by synchrotron X-ray diffraction”. *Acta Materialia* 59.6 (2011), pp. 2501–2513.

[2] V. A. Sharafutdinov. *Integral geometry of tensor fields*. VSP, 1994.

[3] C. Schroer, M. Kuhlmann, S. Roth, et al. “Mapping the local nanostructure inside a specimen by tomographic small-angle x-ray scattering”. *Applied physics letters* 88.16 (2006), p. 164102.

[4] F. Schaff, M. Bech, P. Zaslansky, et al. “Six-dimensional real and reciprocal space small-angle X-ray scattering tomography”. *Nature* 527.7578 (2015), pp. 353–356.

[5] J. Feldkamp, M. Kuhlmann, S. Roth, et al. “Recent developments in tomographic small-angle X-ray scattering”. *physica status solidi (a)* 206.8 (2009), pp. 1723–1726.

[6] A. K. Louis. “Orthogonal function series expansions and the null space of the Radon transform”. *SIAM journal on mathematical analysis* 15.3 (1984), pp. 621–633.

[7] P. Maass. “The x-ray transform: singular value decomposition and resolution”. *Inverse problems* 3.4 (1987), p. 729.

[8] F. Natterer. “The mathematics of computerized tomography (classics in applied mathematics, vol. 32)”. *Inverse Problems* 18 (2001), pp. 283–284.

[9] F. John. “The ultrahyperbolic differential equation with four independent variables”. *Duke Mathematical Journal* 4.2 (1938), pp. 300–322.

[10] W. R. Lionheart and P. J. Withers. “Diffraction tomography of strain”. *Inverse Problems* 31.4 (2015), p. 045005.

[11] N. M. Desai and W. R. B. Lionheart. “An explicit reconstruction algorithm for the transverse ray transform of a second rank tensor field from three axis data”. *Inverse Problems* 32.11 (2016), p. 115009.

[12] M. Liebi, M. Georgiadis, A. Menzel, et al. “Nanostructure surveys of macroscopic specimens by small-angle scattering tensor tomography”. *Nature* 527.7578 (2015), pp. 349–352.

[13] M. Guizar-Sicairos, M. Georgiadis, and M. Liebi. “Validation study of small-angle X-ray scattering tensor tomography”. *Journal of Synchrotron Radiation* 27.3 (2020).

[14] S. Axler, P. Bourdon, and R. Wade. *Harmonic function theory*. Vol. 137. Springer Science & Business Media, 2013.

[15] A. Defant and S. Schlüters. “Non-symmetric polarization”. *Journal of Mathematical Analysis and Applications* 445.2 (2017), pp. 1291–1299.

[16] J. Jørgensen et al. “Core Imaging Library - Part I: a versatile Python framework for tomographic imaging”. *Phil. Trans. R. Soc. A* (submitted) (2021).

[17] C. Kübel, A. Voigt, R. Schoenmakers, et al. “Recent advances in electron tomography: TEM and HAADF-STEM tomography for materials science and semiconductor applications”. *Microscopy and Microanalysis* 11.5 (2005), p. 378.

[18] R. Leary, Z. Saghi, M. Armbrüster, et al. “Quantitative high-angle annular dark-field scanning transmission electron microscope (HAADF-STEM) tomography and high-resolution electron microscopy of unsupported intermetallic GaPd2 catalysts”. *The Journal of Physical Chemistry C* 116.24 (2012), pp. 13343–13352.

[19] S. G. Kazantsev and A. A. Bukhgeim. “Singular value decomposition for the 2D fan-beam Radon transform of tensor fields”. *Journal of Inverse and Ill-posed Problems* 12.3 (2004), pp. 245–278.

[20] V. P. Krishnan, R. K. Mishra, and S. K. Sahoo. “Microlocal inversion of a 3-dimensional restricted transverse ray transform of symmetric *m*-tensor fields”. *arXiv preprint arXiv:1904.02812* (2020).

[21] W. Treimer. “Neutron refraction and small-angle scattering tomography”. *Advanced Tomographic Methods in Materials Research and Engineering* 66 (2008), p. 425.