Pygix: a general python library for fast reduction of 2D grazing-incidence and fibre X-ray scattering data

T. G. Dane,*a*\* S. Lilliu*b* and J. Kieffer*a*

Grazing-incidence X-ray scattering techniques provide a wealth of information on the structuring of thin films and surfaces, applicable to a wide range of physical, chemical and biological systems. We have re-evaluated the remapping of raw detector pixels into reciprocal space maps, paying close attention to generality of detector and sample geometries, and have found that the equations commonly used are only an approximation. A python library has been written to perform remapping of images into reciprocal space and line-profile extraction with accurate intensity conservation. This library is designed to be truly generic, to account for any experimental situation. The transformation and reduction algorithms utilize previously available image remapping libraries that can be run on the GPU allowing extremely fast data processing, typically 3000 images (4 Mpix) per minute on a standard modern computer.

# Introduction

Grazing-incidence X-ray scattering techniques (GIXS) are a valuable tool for probing the electron density of thin films on surfaces or at buried interfaces. The low incident angle (often below the critical angle of total external reflection) allows the suppression of any scattering signal from the substrate thus allowing the extraction of structural parameters thin samples (even down to sub-monolayer) that would otherwise not be detectable in standard transmission experiments. In the small-angle case (GISAXS) correlation and distributions of islands or particles on surfaces, in the wide-angle, diffraction etc. However, the fixed incident angle gives rise to missing data in the out-of-plane direction due to the intersection of the Ewald sphere with the detector. In the wide-angle case this distortion can be extreme (see Fig. x a). Therefore to extract accurate information on the positions of reflections, images must be projected from the detector plane onto a reciprocal space map (see Fig x b). Furthermore, tilted detectors can introduce additional distortions to the data.

Discussion of the projection to reciprocal space. Fixed incident angle. Fibre texture, 2D powder. Polyani, Stribeck. Smilgies calculations, approximation, originally devised for point detectors, not perfectly accurate. Certainly valid in the small-angle approximation but not at very large angles. In fact grazing and fibre are equivalent and should be treated as such.

Whilst a number of software packages have been developed for analysing GIXS data (in both the wide-angle and small-angle regimes), these have a number of drawbacks. Many, require the use of proprietary software (MATLAB, IgorPro), they are focussed on particular data formats or beamlines, require a lot of information, are very slow. Python is a versatile and freely available programming language with a large and active community. Kieffer *et al.* have developed pyFAI, a versatile python library for the reduction of 2D X-ray scattering data in the transmission geometry, which allows the projection of diffraction patterns in polar coordinates (*i.e.* Intensity *vs.* *q*, **) and reduction to 1D line profiles. Here a Python library has been developed, based on the intensity resampling algorithms of pyFAI for 2D diffraction data collected under the grazing-incidence geometry. The library is truly generic in that any experimental

Generic for any grazing and fibre. What about transmission grazing?



**Figure 1** Construction of the wavevector space for GIXS experiments. The incident beam impinges on the sample at angle **i with wavevector **k**i (dashed red lines). The diffracted beam leaves the surface with exit wavevector **k**f (solid red lines) with an in-plane scattering angle of 2**f and out-of-plane angle **i. The total wavevector transfer **q** (solid blue lines) is given by **k**f – **k**i. The individual Cartesian components of each wavevector are shown.

# Geometry

## Detector geometry

In some experiments, detectors may be tilted, either to access a larger region of reciprocal space or simply due to limitations in accurate detector mounting. Such detector tilt imparts a distortion on the resulting diffraction patterns and the detector rotation angles must be taken into account when calculating the reciprocal space coordinates of pixels. Pygix uses the same detector geometrical construction as used by pyFAI, first described by P. Boesecke.[[1]](#endnote-1) The frame of reference is described by the three-dimensional Cartesian axes *x*1 (*e.g.* the *y*-axis of the detector image), *x*2 (*e.g.* the *x*-axis of the detector image) and *x*3 which points along the incoming X-ray beam. We define a position on the detector, which is normal to the sample, the point-of-normal-incidence (PONI). A vector **p** describes the position of a pixel on the detector. The first and second elements, *p*1 and *p*2 are the physical distances (in meter) along the *y*- and *x*-axes of the detector from the position of the PONI. The third element, *p*3 is the physical distance (in meter) from the sample to the PONI:

|  |  |  |
| --- | --- | --- |
|  |  | (1) |

In the case of a detector oriented normal the incoming beam, the PONI is equal to the direct beam position on the detector and the vector **p** describes the pixel positions in the laboratory frame, which can be used to directly calculate the reciprocal space coordinates. If, however, the detector is tilted, the PONI and direct beam are not equivalent. We define three rotation matrices **R**1, **R**2 and **R**3,which describe the detector rotations of angles **1, **2 and **3:

|  |  |  |
| --- | --- | --- |
|  |  | (2) |
|  |  | (3) |
|  |  | (4) |

The reader may question the necessity of the third detector rotation (about the beam axis). Indeed the determination of this angle through fitting observed Debye Scherrer rings of diffraction from a calibration standard is non-trivial, and in most situations this angle is zero. One example of the importance of taking into account this detector rotation however, is the situation in which a detector is placed close to the sample to collect the wide-angle signal but tilted by ~45° such that the beam passes the detector to a second further back in a simultaneous GISAXS-GIWAXS experiment. Finally the laboratory coordinate of the pixel **p**' is given as by the product of the pixel vector **p** with the rotation matrices:

|  |  |  |
| --- | --- | --- |
|  |  | (5) |

Determination of the parameters *l*SD, the PONI and the detector rotation angles are most easily determined by recording a diffraction pattern of a calibration standard (*e.g.* silver behenate, LaB6 or corundum) in transmission mode. PyFAI offers a programme to fit and refine these parameters (*pyFAI-calib*). The fitted parameters are stored in a text file with extension .poni. This file can be passed to pygix or alternatively, the values can be passed directly when instantiating the class.

## Sample geometry

Grazing-incidence experiments can be performed in a number of geometries. Most commonly, the sample is placed horizontally, parallel to the floor of the laboratory (in the *xy* plane). In this situation one can define *qz* as increasing positively from the bottom to the top of the detector image. Alternatively the sample can be mounted normal to the laboratory plane in the vertical scattering geometry and *qz* is along the *x*-axis of the detector image (note the direction of *qz* then depends from which side the incident beam impinges on the sample). Lastly, a less common geometry is that of the surface horizontal but upside-down in which the beam approaches from the underside, for example in studies of hanging drops on surfaces.[[2]](#endnote-2) Thus we define four sample geometries: 1, surface horizontal, *qz* points positive along the image *y*-axis; 2, surface vertical, *qz* points positive along the image *x*-axis; 3, surface horizontal, *qz* points negative along the image *y*-axis; 4, surface vertical, *qz* points negative along the image *x*-axis. Pygix allows for specification of each of these sample geometries.

Two additional sample angles must then be taken into account. Most importantly, the incident angle (**i) and secondly the tilt angle of the surface plane about the beam axis. Ideally, when mounted on a multi-axis goniometer the tilt angle of the sample can be corrected during sample alignment. In certain cases however, either motorisation does not allow for this or data maybe collected erroneously without correcting this angle. The incident angle is required for pygix calculations, the tilt angle is an optional additional parameter (defaults to zero).

# Reciprocal space projection

## Wavevector space

Raw images recorded by the detector represent slices through the intersection of the Ewald sphere with the Polyani sphere. In order to extract quantitative information from these images it is necessary to project these images into relevant coordinate systems. There are three main coordinate systems of interest: angular coordinates (2**f, **f, more frequently used by the GISAXS community), wavevector transfer coordinates (or reciprocal space maps, *qxy*, *qz*, favoured by the GIWAXS/GIXD community but also used by the GISAXS community) and lastly the polar projection, *i.e.*, the absolute wavevector transfer and azimuthal angle (*q*, **), sometimes referred to as cake plots.

For the calculation of reciprocal space coordinates, we define a different coordinate system to that of the calculation of Cartesian pixel coordinates described in Section 2.1. The *x*-axis points along the beam, the *y*-axis, along with the *x*-axis defines the sample surface plane and the *z*-axis is normal to the sample surface. The conversion between the two coordinate systems is described as:

|  |  |  |
| --- | --- | --- |
|  |  | (6) |

First we consider the reciprocal space projection as if the incident and tilt rotation angles of the sample were zero. The wavevector transfer **q** for elastic X-ray scattering is defined as the outgoing wavevector *k*f minus the incoming wavevector *k*i.[[3]](#endnote-3) In this situation, the incoming wavevector *k*i0 is along the incident beam. The expression for the outgoing wavevector, as described by Stribeck, is defined as:[[4]](#endnote-4)

|  |  |  |
| --- | --- | --- |
|  |  | (7) |
|  |  | (8) |
|  |  | (9) |

where the wavenumber (defining the radius of the Ewald sphere) . The individual *x, y* and *z* components of **q**0 are thus defined as:

|  |  |  |
| --- | --- | --- |
|  |  | (10) |
|  |  | (11) |
|  |  | (12) |

We then consider the case where the sample has a non-zero incident angle (or fibre tilt angle) and construct a rotation matrix about the *y*-axis.

|  |  |  |
| --- | --- | --- |
|  |  | (13) |

If the sample is tilted about the beam axis, *i.e.*, the surface plane (or fibre axis) by an angle **, we define a second rotation matrix about the *x*­-axis:

|  |  |  |
| --- | --- | --- |
|  |  | (14) |

We note here, that the sample geometry is taken into account in this rotation matrix. For the standard GIXS geometry, no modification is required; for geometries 2, 3 and 4, values of /2,  and 3/2, respectively, are added to **. The un-corrected wavevector transfer **q**0 is then rotated by these matrices to determine the corrected wavevector transfer, **q**:

|  |  |  |
| --- | --- | --- |
|  |  | (15) |

Finally, by expanding Equation (15) we arrive at the *x, y* and *z* components correct wavevector transfer **q**:

|  |  |  |
| --- | --- | --- |
|  |  | (16) |
|  |  | (17) |
|  |  | (18) |

Due to the in-plane isotropy, GIXS images are typically presented as a function of the out-of-plane (*qz*) and in-plane (*qxy*) wavevector transfer components, where *qxy* is given by:

|  |  |  |
| --- | --- | --- |
|  |  | (19) |

From **q** we derive the polar coordinates, *i.e.* the absolute length of **q** and the azimuthal angle relative to the surface normal (*q*, **):

|  |  |  |
| --- | --- | --- |
|  |  | (20) |
|  |  | (21) |

## Angular space

Referring to Figure 1, one can see that the in-plane scattering angle (2**f) is the angle between *k*f*x* and *k*f*y*,and the out-of-plane scattering angle (**f) is the angle between *k*f*xy* and *k*f*z*. Thus to calculate the scattering angles, we multiply the uncorrected (for sample rotations) exit wavevector, **k**f0 (given by Equation (8)) by the sample rotation matrices, **R***y*(**i)**R***x*(**) (given by Equations (13) and (14)). The Cartesian lengths of the corrected **k**fare then given by:

|  |  |  |
| --- | --- | --- |
|  |  | (22) |
|  |  | (23) |
|  |  | (24) |
|  |  | (25) |

From **k**f the out-of-plane (**f) and in-plane (2**­f) scattering angles are related through Equations (26) and (27):

|  |  |  |
| --- | --- | --- |
|  |  | (26) |
|  |  | (27) |

From the scattering angles we derive the polar coordinates, *i.e.* the absolute scattering angle 2** and the azimuthal angle, ** as follows:

|  |  |  |
| --- | --- | --- |
|  |  | () |
|  |  | () |

# Intensity remapping algorithm

Histogram approach. Pixel splitting. Projection is not surjective.

# Data corrections

In the development of pygix we have tried to consider all possible intensity corrections to the data. Flat field, dark current and mask files can be supplied. These corrections can be classified into two types: corrections applied to the raw images, independent of sample geometry and pixel-wise corrections that are dependent on the experimental geometry.

## Geometry-independent corrections

In recent years the use of single-photon counting detectors has become increasingly popular at synchrotron radiation facilities. For such detectors, counts on the detector are only recorded for photons hitting the detector (though with an efficiency dependency). However, CCD cameras are still widely used. These cameras typically have a dark current noise (*i.e.* the intensity recorded in an image with no exposure to X-rays) and variations in the sensitivity of individual pixels (flat field sensitivity, which can be determined by illuminating the detector with a uniformly radiating fluorescent source). Users can supply dark-current and flat field images when using pygix. For detectors with fibre optic taper distortion (*e.g.* the ESRF FReLoN camera), a spline file can be supplied and the pin cushion distortion will be corrected. Lastly, mask files can be supplied. Whilst masks are dependent on the experimental setup, the mask file describes the regions of the raw image that contain usable data and thus no further calculation is required.

## Geometry-dependent corrections

X-ray photons scattered at different angles have a different path length from the sample to detector, thus have a greater absorption due to the medium at greater scattering angles. In the case of small-angle scattering this absorption variation is negligible and vacuum flight tubes are often employed to minimise absorption. For experiments in air at very short detector distances, however, this variation can be significant (of the order of a few %). Whilst it has previously been stated that this depends on the absolute scattering angle 2**, this is only valid for detectors positioned normal to the incident beam. Correction for absorption by the medium is therefore based on the physical distance of each sample to the detector in pygix using the following equation:

|  |  |  |
| --- | --- | --- |
|  |  | (28) |

where **m is the absorption coefficient of the medium and *d* is the distance from the pixel to the sample. Similarly photons hitting the detector at lower incident angles experience longer path lengths through the detector, thus greater absorption and a higher probability of the photon being detected. As for the medium absorption correction, for tilted detectors, this term should not be calculated from the scattering angle but from the incident angle of the photons on the detector (**). In pygix this is calculated as:

|  |  |  |
| --- | --- | --- |
|  |  | (29) |

where **d and *t*d are the absorption coefficient and thickness of the detector, respectively.

The area of reciprocal space subtended by a pixel on the detector is also dependent on the incident angle of photons on the detector. The greatest area of reciprocal space is observed by the pixels closest to the PONI. As photons are scattered further away from the PONI each pixel sees less of reciprocal space and thus less intensity.

To discuss:

* Polarization.
* Lorentz correction.
* Jacobian determinant of the fibre projection.
* Sample absorption, illumination area, normalization factor. Illumination area depends on omega and incident angle. Handled from the user side, provide one value to normalization factor. i.e. *I*0 = 73 % of maximum, sample sees 53 % of incoming intensity normalization factor = (0.53\*0.73), pygix divides intensity map by normalization factor
* Mention background subtraction – difficult. If user wants to do process in the same way taking care to normalize and subtract from data.

# Summary of the main pygix methods

## 2D image projections

## 1D data reduction

## Additional scripts

Sample alignment, batch processing.

# Conclusions

Develop an open source library independent of data format, truly generic for all data types

# Acknowledgements

TGD is extremely grateful for helpful discussions with M. Burghammer, C. Riekel, B. Weinhausen, M. Rosenthal and J. D. Nicholas. TGD and SL are greatly indebted to J. E. Macdonald for many helpful discussions and for reading through the manuscript. We wish to thank all of the contributors to the pyFAI package, without which this package would not have been possible.

1. Boesecke, P. (2007). Reduction of two-dimensional small- and wide-angle X-ray scattering data. *Journal of Applied Crystallography*, *40*(s1), s423–s427. doi:doi:10.1107/S0021889807001100 [↑](#endnote-ref-1)
2. Pechkova, E., Gebhardt, R., Riekel, C., & Nicolini, C. (2010). In situ microGISAXS: I. Experimental setup for submicron study of protein nucleation and growth. *Biophysical Journal*, *99*(4), 1256–1261. doi:10.1016/j.bpj.2010.03.069 [↑](#endnote-ref-2)
3. Als-Nielsen, J., & McMorrow, D. (2011). *Elements of Modern X-Ray Physics* (2nd ed.). Chichester, United Kingdom: John Wiley & Sons Ltd. [↑](#endnote-ref-3)
4. Stribeck, N., & Nöchel, U. (2009). Direct mapping of fiber diffraction patterns into reciprocal space. *Journal of Applied Crystallography*, *42*(2), 295–301. doi:10.1107/S0021889809004713 [↑](#endnote-ref-4)