# Processing scanning Laue microdiffraction patterns with machine learning algorithms: A case-study with fatigued polycrystalline copper

## Introduction

Laue diffraction, that may occur when a polychromatic X-ray beam illuminated a crystal, was first discovered in 1912, and has revealed both the electromagnetic nature of X-ray and the periodic ordering of atoms in crystal [1]. Thanks to the polychromaticity of the employed X-ray, multiple diffraction peaks can be recorded in a single exposure without any rotation that might lead to the ambiguity of the illuminated volume [2]. With the development of polychromatic beam focusing optics, notably Kirkpatrick–Baez mirrors, micron-sized high-brilliance polychromatic X-ray beam can be produced at synchrotron radiation sources and directed to probe inside materials with submicrometric spatial resolution, i.e. Laue microdiffraction. In analogy to EBSD (electron backscatter diffraction) technique, Laue microdiffraction technique serves by raster scanning the sample to generate the lattice orientation and distortion maps from the one-by-one analysis of the diffraction pattern emanating from each scanned spot [3-5]. The two techniques are comparable [6] and complementary to each other [7]. It is generally accepted that EBSD has an edge on finer spatial resolution of nanoscale, whilst Laue microdiffraction can have a much better accuracy on the lattice orientation and distortion with an attainable order of 10-9 [8].

A salient feature of Laue microdiffraction is its sensitivity to the local misorientation inside the illuminated volume [9, 10], more specifically, the fragmentation of Laue spot may indicate the presence of geometrically necessary boundaries and the elongation of Laue spot the presence of geometrically necessary dislocations. Although a critical aspect of the spot shape analysis lies on the assumption that the dislocations were dominantly edge-type in the illuminated volume, a recent study with focused ion beam transmission electron microscopy confirmed that this analysis stood still if the dislocations had predominately screw-type [11]. With the aid of a wire profiler (typically Pt), the shape of spot can be spatially resolved to yield subsurface, 3D mapping of lattice orientation and distortion non-destructively [12-14], namely the differential-aperture X-ray microscopy (DAXM) technique [15].

Despite the wealth of information behind Laue microdiffraction pattern, the interpretation of Laue microdiffraction pattern is not straightforward since the wavelength or indexation pertaining to each diffraction peak is unknown. Standard treatment involves modulating the orientation and calibration parameters to minimize the discrepancy between the simulated and experimental diffraction pattern, and has been implemented in software such as *XMAS* [16] and *LaueTools* (<https://gitlab.esrf.fr/micha/lauetools>). The standard treatment is in essence a trial-and-error process that usually suffers from inefficiency, especially for the raster scanned diffraction patterns which has to be treated one by one. Therefore, any additional information concerning the scanned microstructure would facilitate the process, for example, Örs et al. [7] used the orientation obtained by EBSD to overcome the difficulty in indexing the Laue microdiffraction patterns; Kou et al. [17] suggested indexing one Laue microdiffraction pattern per grain as the reference with which the rest patterns of the grain could be analyzed without indexation.

Concurrently, the emerging image processing techniques have demonstrated their potential in the interpretation of Laue microdiffraction patterns: Petit et al. [18] introduced the digital image correlation (DIC) technique to have a better measurement of relative lattice distortion with reference to an assumed stress-free position, i.e. Laue-DIC method, Zhang et al. [19] later extended the framework of Laue-DIC to get rid of the dependence on the stress-free reference; Zhang et al. [20] used DIC to correct the misalignment of the investigated volume in the experiments of DXAM;

In a word, the indexation of diffraction peaks is the key to full interpretation of diffraction pattern. However, in certain circumstances, full interpretation of diffraction patterns is unnecessary or fast parallel computing capabilities are unavailable, thereby necessitating the development of indexation-free approach towards on-the-fly analysis of raster scanned diffraction patterns. Zhou et al. [5] proposed using the distribution of average recorded intensities and average filtered intensities of the raster scanned diffraction patterns to visualize the characteristics microstructural features. The recently emerging convolutional neural networks (CNN) has been used to extract features from diffraction patterns for further clustering and labeling the raster scanned diffraction patterns [21].

In the present work, we demonstrated the application of machine learning algorithms to the raster scanned diffraction patterns of the fatigued polycrystalline copper. Substantial dislocation structures will grow in copper after the cyclic loading [22], deteriorating the identifiability of the diffraction pattern. Although template matching schemes have been shown applicable to the indexation and misorientation analysis of smeared diffraction patterns, huge amount of calculation was still inevitable [23] and the reliability of outcome would be degraded in line with the formation of dislocation structures. On the other hand, machine learning algorithms, which were developed to handle big data, were possible to circumvent the difficulty of indexation and segment raster scanned diffraction patterns according to their features, thereby mapping phases, grains, or grain substructures. Since the microstructure of the scanned area was not known *a priori*, unsupervised labeling algorithms had to be adopted. The objective of this paper is to: (i) outline the computational pipeline from the raw data to the clustering of diffraction patterns; (ii) compare the performance of several algorithms; and (iii) discuss the influence of the diffraction patterns on the results of clustering.

## Experiment

The diffraction patterns were collected from raster scanning of fatigued polycrystalline copper. The sample was designed in accordance with the ASTM/E606 standard. The tensile strength at room temperature was given in Fig 1 along with the EBSD (electron backscattered diffraction) mapping in the inset. The sample, cyclically loaded in stress-control mode with the stress varying sinusoidally within the range 0 ~ 140 MPa, has undergone a maximum strain of ~ 10% in the initial cycle from Figure 1 and cyclic creep in the subsequent cycles. The sample was fatigued up to 109 cycles with a frequency of 10 Hz.



Figure 1 The tensile curve of the sample along with the orientation mapping in the inset.

The Laue raster scanning over the sample was performed in beamline 4B of Pohang Light Source with a step size of 2 μm. The obtained Laue microdiffraction pattern was extremely blurred with almost no discernable diffraction peaks (Figure 2a).

## Data reduction

The original Laue microdiffraction pattern has 1024×1024 pixels (Figure 2a). It is impractical to handle such huge amount of data, thus a data reduction process is necessary to reduce the diffraction patterns into a manageable number of latent features. To begin with, each image needed to be normalized to eliminate systematic errors. Normalization was accomplished by subtracting the mean gray level from the gray level and dividing by the standard deviation of the gray levels (Figure 2a). Then the normalized images were shrunk to 128×128 pixels by 8×8 averaging binning to reduce the size of data and smooth the noise (Figure 2b). The images in spatial domain could be equivalently expressed in frequency domain (Figure 2c) by discrete sine transformation (DST), which transformed the image into the weighted sum of sinusoids with discrete frequencies. It was obvious from Figure 2c that the components of high frequencies were negligible compared to those of low frequencies.



Figure 2 (a) The normalized image of the diffraction pattern; (b) the shrunk image of the Figure 2a after 8×8 averaging binning; (c) the DST of Figure 2b.

Both the images in spatial and frequency domain had 128×128 pixels for clustering. Song et al. applied CNN to extract latent features [21]. However, at present, the authors did not have sufficiently large date set to train the CNN, therefore unsupervised learning algorithms that did not require labeling of data were employed herein to extract latent features of each image. If the latent features were properly extracted, it was possible to classify the diffraction patterns to the grains which they belonged to. Here we used the hierarchical agglomerative clustering (HAC) algorithm backed by Scikit-Learn [24]. When treating the scanning diffraction images with HAC algorithm, each pixel corresponded to a vector comprised of the values at the pixel in all images whether they be in spatial or frequency domain; then a metric (Euclidean distance, maximum distance, etc.) was used to quantify the dissimilarity between the pairs of pixels; then pixels with high similarities were merged to form a feature according to a linkage criterion. The connectivities of pixels could be exploited to facilitate the merging process such that only the pairs of adjacent pixels were under consideration. In this work, 256 latent features were to be extracted from the shrunk images wherein the Euclidean distance was used as the metric of dissimilarity, and the linkage criterion employed was “ward” aiming at minimizing the sum of squared differences within all clusters.

Figure 2d and e display the results of feature clustering of Figure 2b and c respectively, and Figure 2f plots the histograms of the numbers of pixels contained by each feature in both spatial and frequency domain. The feature clustering was rather uniform in the spatial domain whilst the features concentrated on the lower frequencies in the frequency domain. A highly skewed distribution of pixel numbers in frequencies domain was identified, suggesting that the majority of high frequency components could be merged into one feature, whereas the pixel numbers distributed uniformly among features.

[1] M. Eckert, Disputed discovery: the beginnings of X-ray diffraction in crystals in 1912 and its repercussionsThis Laue centennial article has also been published in Zeitschrift fur Kristallographie [Eckert (2012). Z. Kristallogr. 227, 27-35], Acta Crystallogr. Sect. A 68(1) (2012) 30-39.

[2] J.-S. Chung, G.E. Ice, Automated indexing for texture and strain measurement with broad-bandpass x-ray microbeams, J. Appl. Phys. 86(9) (1999) 5249-5255.

[3] R. Spolenak, W.L. Brown, N. Tamura, A.A. MacDowell, R.S. Celestre, H.A. Padmore, B. Valek, J.C. Bravman, T. Marieb, H. Fujimoto, B.W. Batterman, J.R. Patel, Local Plasticity of Al Thin Films as Revealed by X-Ray Microdiffraction, Phys. Rev. Lett. 90(9) (2003) 096102.

[4] N. Tamura, A.A. MacDowell, R. Spolenak, B.C. Valek, J.C. Bravman, W.L. Brown, R.S. Celestre, H.A. Padmore, B.W. Batterman, J.R. Patel, Scanning X-ray microdiffraction with submicrometer white beam for strain/stress and orientation mapping in thin films, J. Synchrotron Rad. 10(2) (2003) 137-143.

[5] G. Zhou, J. Kou, Y. Li, W. Zhu, K. Chen, N. Tamura, Quantitative Scanning Laue Diffraction Microscopy: Application to the Study of 3D Printed Nickel-Based Superalloys, Quantum Beam Sci. 2(2) (2018) 13.

[6] E. Plancher, J. Petit, C. Maurice, V. Favier, L. Saintoyant, D. Loisnard, N. Rupin, J.B. Marijon, O. Ulrich, M. Bornert, J.S. Micha, O. Robach, O. Castelnau, On the Accuracy of Elastic Strain Field Measurements by Laue Microdiffraction and High-Resolution EBSD: a Cross-Validation Experiment, Exp. Mech. 56(3) (2016) 483-492.

[7] T. Örs, J.-S. Micha, N. Gey, V. Michel, O. Castelnau, R. Guinebretiere, EBSD-assisted Laue microdiffraction for microstrain analysis, J. Appl. Crystallogr. 51(1) (2018) 55-67.

[8] C. Zhang, T.R. Bieler, P. Eisenlohr, Exploring the accuracy limits of lattice strain quantification with synthetic diffraction data, Scr. Mater. 154 (2018) 127-130.

[9] R. Barabash, G.E. Ice, B.C. Larson, G.M. Pharr, K.-S. Chung, W. Yang, White microbeam diffraction from distorted crystals, Appl. Phys. Lett. 79(6) (2001) 749-751.

[10] R.I. Barabash, G.E. Ice, F.J. Walker, Quantitative microdiffraction from deformed crystals with unpaired dislocations and dislocation walls, J. Appl. Phys. 93(3) (2003) 1457-1464.

[11] C. Zhang, S. Balachandran, P. Eisenlohr, M.A. Crimp, C. Boehlert, R. Xu, T.R. Bieler, Comparison of dislocation content measured with transmission electron microscopy and micro-Laue diffraction based streak analysis, Scr. Mater. 144 (2018) 74-77.

[12] W. Yang, B.C. Larson, J.Z. Tischler, G.E. Ice, J.D. Budai, W. Liu, Differential-aperture X-ray structural microscopy: a submicron-resolution three-dimensional probe of local microstructure and strain, Micron 35(6) (2004) 431-439.

[13] R.I. Barabash, G.E. Ice, W. Liu, O.M. Barabash, Polychromatic microdiffraction characterization of defect gradients in severely deformed materials, Micron 40(1) (2009) 28-36.

[14] S. Das, F. Hofmann, E. Tarleton, Consistent determination of geometrically necessary dislocation density from simulations and experiments, Int. J. Plast. 109 (2018) 18-42.

[15] B.C. Larson, W. Yang, G.E. Ice, J.D. Budai, J.Z. Tischler, Three-dimensional X-ray structural microscopy with submicrometre resolution, Nature 415(6874) (2002) 887-890.

[16] N. Tamura, XMAS: A Versatile Tool for Analyzing Synchrotron X-ray Microdiffraction Data, Strain and Dislocation Gradients from Diffraction2014, pp. 125-155.

[17] J. Kou, K. Chen, N. Tamura, A peak position comparison method for high-speed quantitative Laue microdiffraction data processing, Scr. Mater. 143 (2018) 49-53.

[18] J. Petit, O. Castelnau, M. Bornert, F.G. Zhang, F. Hofmann, A.M. Korsunsky, D. Faurie, C. Le Bourlot, J.S. Micha, O. Robach, O. Ulrich, Laue-DIC: a new method for improved stress field measurements at the micrometer scale, J. Synchrotron Rad. 22(4) (2015) 980-994.

[19] F.G. Zhang, O. Castelnau, M. Bornert, J. Petit, J.B. Marijon, E. Plancher, Determination of deviatoric elastic strain and lattice orientation by applying digital image correlation to Laue microdiffraction images: the enhanced Laue-DIC method, J. Appl. Crystallogr. 48(6) (2015) 1805-1817.

[20] C. Zhang, Y. Zhang, G. Wu, W. Liu, R. Xu, D. Juul Jensen, A. Godfrey, Alignment of sample position and rotation during in situ synchrotron X-ray micro-diffraction experiments using a Laue cross-correlation approach, J. Appl. Crystallogr. 52(5) (2019) 1119-1127.

[21] Y. Song, N. Tamura, C. Zhang, M. Karami, X. Chen, Data-driven approach for synchrotron X-ray Laue microdiffraction scan analysis, Acta Crystallogr. Sect. A 75(6) (2019) 876-888.

[22] H. Mughrabi, Cyclic Slip Irreversibilities and the Evolution of Fatigue Damage, Metall. Mater. Trans. B 40(4) (2009) 431-453.

[23] V.K. Gupta, S.R. Agnew, Indexation and misorientation analysis of low-quality Laue diffraction patterns, J. Appl. Crystallogr. 42(1) (2009) 116-124.

[24] F. Pedregosa, G. Varoquaux, A. Gramfort, V. Michel, B. Thirion, O. Grisel, M. Blondel, P. Prettenhofer, R. Weiss, V. Dubourg, J. Vanderplas, A. Passos, D. Cournapeau, M. Brucher, M. Perrot, É. Duchesnay, Scikit-learn: Machine Learning in Python, J. Mach. Learn. Res. 12(null) (2011) 2825–2830.