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On *ab initio* indexing of Laue diffraction patterns

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The Laue method allows for fast pattern acquisition, but its use in structural studies is limited by the complexity of data processing. In particular, automatic *ab initio* indexing of Laue patterns is not trivial. This paper describes measures improving the effectiveness of indexing software. The first such measure is to adjust the positions of Laue spots on the basis of a mesh of lines fitted in a consistent way. Two other modifications enlarge the set of cells tested as potential primitive lattice cells. The last modification concerns eliminating solutions representing superlattices of the true reciprocal lattice. The impact of using these schemes on the chances of obtaining correct indexing solutions is illustrated. The described procedures can be implemented to create fully automatic software for *ab initio* indexing of Laue patterns.

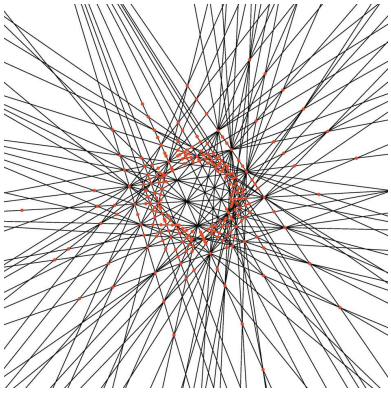
1. Introduction

With Laue diffraction, pattern acquisition times are much shorter than with monochromatic techniques (Amorós *et al.*, 1975). The high speed of data collection is an important advantage in time-resolved studies (Ren *et al.*, 1999; Popov *et al.*, 2019). Moreover, pink-beam serial crystallography has emerged as a promising method for determining the structures of macromolecules (Förster & Schulze-Briese, 2019). The Laue method, however, is not commonly used because of difficulties with data processing (Cornaby *et al.*, 2010; Förster & Schulze-Briese, 2019).

There are numerous accounts of indexing Laue patterns originating from crystals of known lattice parameters. In contrast, literature on *ab initio* indexing is scarce. Carr *et al.* (1992) described indexing via visual recognition of special configurations of spots in the gnomonic projection. Building on that approach, Ravelli *et al.* (1996) developed *LaueCell*, a semi-automatic indexing program. The *XMAS* package for multigrain indexing of Laue patterns and strain refinement (Tamura, 2014) has a routine for *ab initio* indexing based on the algorithm of Ravelli and co-workers.

In the description of the *LaueG* program for processing neutron Laue patterns (Piltz, 2018), *ab initio* indexing is listed among 'capabilities under development'.

Here, some modifications of known indexing procedures are proposed. They improve the effectiveness of the indexing, and the schemes can be of use in cases which require a fully automatic approach (*e.g.* serial crystallography) or when indexing is difficult due to a large fraction of undetected spots.



2. Steps towards automatic indexing

Any reasonably complete introduction to the indexing of Laue patterns would require a much longer text. For brevity, it is assumed that the reader is familiar with the aforementioned

paper (Ravelli *et al.*, 1996). Let us only recall that *ab initio* indexing amounts to the determination of lattice parameters and lattice orientation. Since two lattices differing only by a scale factor lead to identical Laue patterns, the geometry of a Laue pattern can provide only relative dimensions of the lattice primitive cell. Additional information is needed to get the absolute cell dimensions (Carr *et al.*, 1992, 1993). Indexing can be facilitated by using the relationship between the positions of the spots on a Laue pattern and the gnomonic projection of normalized reciprocal-lattice vectors (see *e.g.* Wyckoff, 1920): reflections belonging to the same zone appear on a conic on the pattern, and the endpoints of the corresponding vectors are projected on a straight line.

2.1. On fitting a consistent mesh

In standard practice, peak-search routines are used to get the spot positions on the pattern [see *e.g.* Ravelli *et al.* (1996)], and the distance between the measured and predicted positions is minimized at a later stage of refinement when the approximate lattice parameters are known. The positions of the spots can be corrected at the outset of processing by fitting conics. The problem is linear [see *e.g.* Morawiec (2004)] so the fitting can be easily performed. The use of direct-space vectors corresponding to conics by Ravelli *et al.* (1996) is a form of such fitting.

Alternatively, one can use the relationship between the Laue pattern and the gnomonic projection, and replace the fitting of conics by the fitting of straight lines (only the latter approach will be considered below). The fitting is linked to the key step of ascribing Laue spots to particular zones.

The proposed modification is to fit lines in a consistent way. Generally, n lines may intersect at $n(n-1)/2$ distinct points. However, a diffraction spot ascribed to more than two lines provides additional constraints on the fitted lines as they should intersect at a single point near the spot. Therefore, rather than fitting individual lines sequentially, one can fit them simultaneously using the constraints imposed by the identity of such intersection points. Ultimately, this approach gives spot positions more consistent and better matching to a lattice than those resulting from the application of only a peak-search algorithm. In particular, the positions of points at the intersections of lines can be given with the accuracy of numerical processing, and (with the same accuracy) they match a certain reciprocal lattice. In other words, by fitting a consistent mesh, one partly refines the projection of the lattice without knowing the lattice parameters, and the refined part of the projection practically determines the lattice.

2.2. On using sextuplets of points

In *ab initio* indexing, the key task is to obtain the parameters of a primitive cell of the crystal lattice. With the centre of the projection at a cell vertex, the projection of the remaining vertices on the unit sphere is a septuple of specially arranged points (Amorós *et al.*, 1975). The approach of Carr *et al.* (1992) and Ravelli *et al.* (1996) is to search for such septuplets.[1] However, some spots are missed in automatic detection and some are absent, and the septuplets are scarce. The problem can be addressed by noting that the cell is actually determined by a smaller number of spots. Instead of looking for septuplets, one can get a cell from a smaller number of spots and test whether a basis ascribed to the cell indexes other spots of the pattern. These can be specially arranged sextuplets (Fig. 1), and clearly one could also get cells from quintuplets or quadruplets of points. Using such smaller configurations significantly increases the number of tested cells and the likelihood of encountering the correct one.

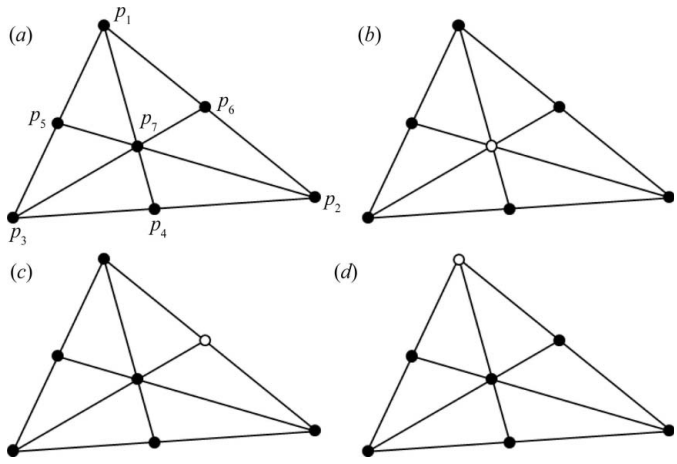


Figure 1
Illustration of projections of a parallelepipedal cell from one of its vertices. In general, the remaining vertices are projected onto seven specially arranged points p_1, p_2, ..., p_7 shown in (*a*). The (relative) cell parameters are determined by a smaller number of points, in particular by sextuplets of types shown in (*b*)–(*d*). Points marked by open circles are missing.

An incomplete configuration can be used for additional peak detection. Having, say, a sextuplet of spots, the software can compute the coordinates of the seventh point and go back to the diffraction pattern to check for the existence of the missing spot. Similarly, with the mesh described in Section 2.1, having a line intersection, one can check for a missing Laue spot.

2.3. On testing superlattices

A cell resulting from the previous steps is likely to be a multiple of a primitive cell. To increase the chances of encountering primitive cells, Ravelli *et al.* (1996) used 'main' conics, *i.e.* conics bordered by wide clear regions and having a small average spacing between the spots. But there is an alternative approach. The obtained reciprocal-space cell may be primitive with respect to a sublattice of the true reciprocal lattice. Therefore, to increase the probability of finding the true primitive cell, besides testing the reciprocal-lattice basis spanning a given cell, one can also test the bases of low-index superlattices of that lattice. This approach is practically the same as that applied in the program *Ind_X* for indexing data collected using monochromatic techniques (Morawiec, 2017).

[1] Ravelli *et al.* (1996) search for the septuplets in direct space. Here, all considerations are limited to reciprocal space.

A method of generating superlattices is described by Santoro & Mighell (1973).

2.4. On the figure of merit

Clearly, if a diffraction pattern can be indexed using a given reciprocal lattice, it can also be indexed using its superlattices. In conventional indexing of single-crystal patterns, when the magnitudes of the scattering vectors are known, if the actual reciprocal lattice is a proper sublattice of the predicted lattice, at least one index of all reflections is a multiple of an integer larger than 1. For instance, if the true basis vector $\mathbf{a}_3^*$ equals two times the predicted vector, then all indices l ascribed to legitimate reflections will be even. This allows for the easy automatic identification and elimination of such cases. In Laue

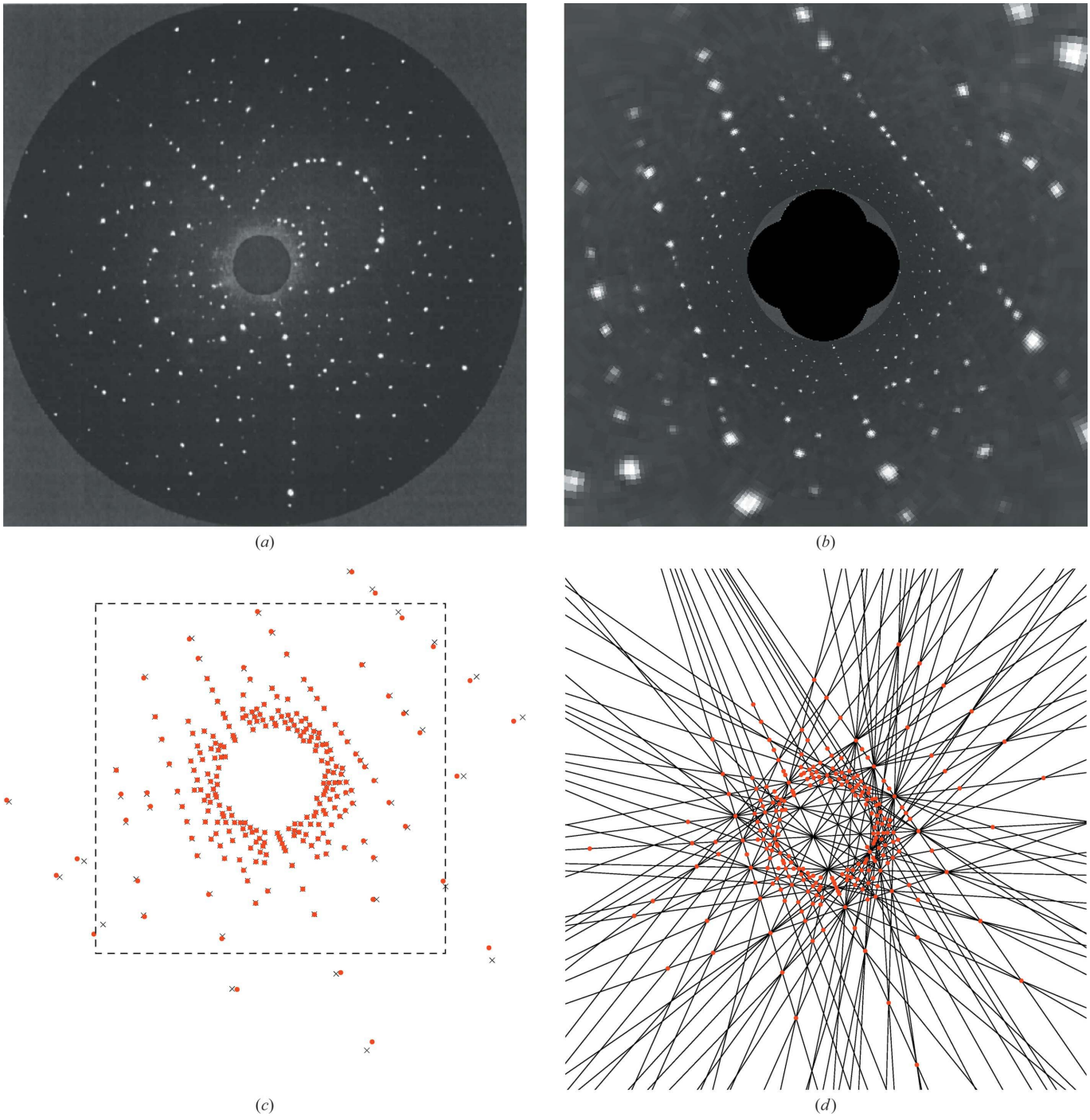


Figure 2
(*a*) Experimental Laue pattern of an organic compound published by Ravelli *et al.* (1996). Reproduced with permission of the International Union of Crystallography. (*b*) Part of the pattern transformed to a gnomonic projection. (*c*) Projection of detected spots (black crosses) and their final positions (red circles) matching the mesh of lines. The dashed square marks the approximate location of the image shown in panel (*b*). (*d*) Each line of the mesh was fitted to at least seven points.

diffraction, the magnitudes of the scattering vectors are not known, and the indices *hkl* ascribed to the spots are relatively prime even if the true indices of the reflection are not, *i.e.* the actual indices of a reflection are divided by their largest common divisor. This eliminates the aforementioned property; the true reciprocal lattice may be a proper sublattice of the predicted lattice with none of h, k or l being a multiple of a fixed integer (>1) for all reflections.

Therefore, the criterion based on the presence of a common divisor of an index for all reflections needs to be replaced by a mechanism modifying the figure of merit (*i.e.* the quantitative measure of the quality of the solutions) based on the frequency of occurrence of such a divisor. In practice, the key point is to check what the fractions are of odd indices h, k and l for a given solution, and to reduce the solution's figure of merit if there is a large disparity between the fractions.

3. Tests

Fig. 2 illustrates the processing of a pattern taken from Ravelli *et al.* (1996). As a proof-of-concept of consistent fitting, the mesh was created in a simple iterative way: at each step, lines passing near a sufficiently large number of points were determined, they were fitted to these points with a weighting accounting for the projection, and then the positions of the points were moved slightly towards the fitted lines. In the particular case shown in Fig. 2, the root-mean-square deviation of spot positions observed on the 512×512 pixel diffraction pattern from their final positions was 0.42 of a pixel. Of 246 detected spots, 181 were ascribed to line intersections.

A prototype indexing program (a Windows version of the program is available at http://imim.pl/personal/adam.morawiec/) was written to test the impact of the procedures described in Sections 2.2 and 2.3. Example indexing rates (*i.e.* fractions of instances when the correct solution is listed) for septuplets, sextuplets of two types and a number of superlattices are given in Table 1. They were obtained from a set of 100 patterns simulated on the basis of data given in frame number 3 of Ravelli *et al.* (1996) [the frame corresponds to the pattern shown in Fig. 2(*a*)]. The crystal orientations were random, uniformly distributed random errors were added to the spot positions, and N randomly selected spots were used for indexing. The results depend on the simulation parameters (magnitude of errors, largest allowed Miller index) and on the thresholds used in indexing, but for all cases listed in Table 1, the same simulation parameters and thresholds were used. In particular, the largest allowed Miller index in both simulation and indexing was 12.

In the program, the quality of the solutions is initially quantified in the way described by Morawiec (2017), but at the end, to account for superlattices of the true lattice (Section 2.4), a simple quality-modifying factor is used: with n_h, n_k and n_l denoting the number of odd h, k and l indices, respectively, if $x = \min\{n_h, n_k, n_l\}/\max\{n_h, n_k, n_l, 1\} < a = 1/2$, the quality of the solution is reduced by a factor $(2a - x)x/a^2$.

Table 1
Example indexing rates for different numbers of points used to get a cell and different indices of tested superlattices.

N is the number of reflections used in indexing. $M_{k,l}$ means that indexing was performed using k-tuplets and all superlattices up to index l. The symbols 6*b* and 6*c* denote sextuplets of the types shown in Figs. 1(*b*) and 1(*c*), respectively. The data were obtained for simulated patterns; see text for details.

N	$M_{7,1}$	$M_{7,3}$	$M_{7,8}$	$M_{7,16}$	$M_{6b,1}$	$M_{6c,1}$	$M_{6c,3}$	$M_{6c,8}$
90	0.02	0.04	0.14	0.18	0.31	0.13	0.34	0.68
120	0.02	0.14	0.43	0.60	0.57	0.23	0.67	0.96
150	0.12	0.45	0.75	0.91	0.75	0.43	0.89	0.99
180	0.20	0.66	0.93	0.97	0.88	0.66	0.94	1.00

The program was applied to the spots detected on the pattern shown in Fig. 2(*a*). In this case, automatic indexing of all 246 spots gives the correct lattice parameters [close to those listed by Ravelli *et al.* (1996)] even if the simple method $M_{7,1}$ is used. When the program is applied to data modified by fitting a consistent mesh (181 reflections ascribed to intersections of lines), the threshold on the angular deviations between the scattering vectors and reciprocal-lattice vectors can be significantly reduced. Both the original (246 spots) and modified (181 spots) data sets are solved with a threshold of 1°. If, however, the threshold is, say, 0.1°, only the second data set gives the correct solution.

4. Conclusions

The procedures described in this paper improve the effectiveness of automatic indexing of Laue diffraction patterns. The first of the proposed schemes is to adjust spot positions on the basis of a mesh fitted to the spots in a consistent way. It resolves the critical issue of ascribing reflections to particular zones.

Two other amendments increase the number of cells tested as potential primitive cells of the crystal lattice. In previous approaches, the cells were constructed by searching for specially arranged septuplets of Laue spots. The simple modification is to use configurations built of a smaller number of spots, *e.g.* sextuplets. Moreover, the likelihood of detecting a primitive cell can be increased by testing subcells of the reciprocal-space cells based on these special spot configurations.

Clearly, with a larger number of tested cells, the computation time is increased. The time also depends strongly on other factors, *e.g.* the number of reflections and the levels of thresholds used to account for errors in the spot positions. One needs a suitable choice of program parameters to keep the time reasonably short.

The last point concerns eliminating solutions representing superlattices of the true reciprocal lattice. The figure of merit of a solution needs to account for the frequencies of occurrence of odd Miller indices.

Results obtained using the prototype software written to test the described procedures indicate that they significantly increase the chances of reaching the correct indexing solution,

and that indexing can be fully automatic for a range of crystal structures.

References

Amorós, J. L., Buerger, M. J. & de Amorós, M. C. (1975). *The Laue Method.* New York: Academic Press.

Carr, P. D., Cruickshank, D. W. J. & Harding, M. M. (1992). *J. Appl. Cryst.* **25**, 294–308.

Carr, P. D., Dodd, I. M. & Harding, M. M. (1993). *J. Appl. Cryst.* **26**, 384–387.

Cornaby, S., Szebenyi, D. M. E., Smilgies, D.-M., Schuller, D. J., Gillilan, R., Hao, Q. & Bilderback, D. H. (2010). *Acta Cryst.* D**66**, 2–11.

Förster, A. & Schulze-Briese, C. (2019). *Struct. Dyn.* **6**, 064302.

Morawiec, A. (2004). *Orientations and Rotations. Computations in Crystallographic Textures.* Berlin: Springer-Verlag.

Morawiec, A. (2017). *J. Appl. Cryst.* **50**, 647–650.

Piltz, R. O. (2018). *J. Appl. Cryst.* **51**, 963–965.

Popov, D., Velisavljevic, N. & Somayazulu, M. (2019). *Crystals*, **9**, 672.

Ravelli, R. B. G., Hezemans, A. M. F., Krabbendam, H. & Kroon, J. (1996). *J. Appl. Cryst.* **29**, 270–278.

Ren, Z., Bourgeois, D., Helliwell, J. R., Moffat, K., Šrajer, V. & Stoddard, B. L. (1999). *J. Synchrotron Rad.* **6**, 891–917.

Santoro, A. & Mighell, A. D. (1973). *Acta Cryst.* A**29**, 169–175.

Tamura, N. (2014). *Strain and Dislocation Gradients from Diffraction: Spatially Resolved Local Structure and Defects*, edited by R. Barabash & G. Ice, pp. 125–155. London: Imperial College Press.

Wyckoff, R. W. G. (1920). *Am. J. Sci.* **50**, 317–360.