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# On the reliability of fully automatic indexing of electron diffraction patterns obtained in a transmission electron microscope

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Orientation maps similar to electron backscatter diffraction maps can be obtained in a transmission electron microscope. A method of such mapping by automatic indexing of electron diffraction patterns has been proposed recently. The procedure is relatively simple and fast but it does not avoid the 180° ambiguity. Using tests on simulated patterns, it is shown that under current practice automatic indexing may give a considerable fraction of erroneous solutions. Optimization of measurement conditions leading to a reduction of that fraction is considered.

Transmission-electron-microscopy-based electron diffraction crystallography is developing in a number of directions. Besides the mainstream interactive analysis of diffraction patterns for phase identification, or structural investigation, there is an interest in automatic indexing of patterns originating from known simple structures. The ultimate goal is the determination of orientations, which are then used for orientation mapping (Wright & Dingley, 1998; Morawiec *et al.*, 2002; Fundenberger *et al.*, 2003; Zaefferer, 2003). One of the interesting concepts in this field is to obtain orientations from electron diffraction patterns (Bragg peaks) formed with focused beams. (These are similar in appearance to selected-area diffraction patterns, but the latter are formed with parallel illumination.) Such an approach has been taken recently by Rauch & Dupuy (2005). They implemented a simple and clever idea of determining orientations by matching experimental patterns to pre-calculated template patterns.

For a known structure, orientation determination from a single pattern is practically equivalent to (three-dimensional) indexing. Questions about the reliability of indexing of selected area diffraction patterns were raised a long time ago (*e.g.* Ryder & Pitsch, 1967). Owing to the dichotomous nature of the indexing problem (a solution is either fully correct or absolutely wrong), correctness of indexing can be relatively easily verified if a small set of patterns is considered, and an interactive approach is common in indexing programs (see *e.g.* Brink & Tam, 1996; Belletti *et al.*, 2000; Berg *et al.*, 2000; Dimmeler & Schroeder, 2000). However, an orientation mapping requires thousands of patterns, and it is essential that the indexing procedure is fully automatic. There are a number of factors influencing pattern generation; besides the crystal orientation these are the structural parameters and the microscope operating conditions. Therefore, predicting the reliability of the final results of indexing is difficult. The optimal guideline is for a given material, use the microscope operating conditions that guarantee that good quality patterns are solvable for all crystalline orientations, but is this possible in practice? It transpires that, even in the simplest cases, there are orientations that lead to unsolvable patterns.

Most of the problems with orientation determination are caused by the so-called 180° ambiguity with reflections from planes belonging to one low-index zone axis. The issue can be illustrated using the Cu [112] pattern shown in Fig. 1. The zeroth-order Laue zone contains reflections $hkl$ satisfying $h + k + 2l = 0$. This pattern has twofold symmetry with respect to the microscope axis; the rotation about the [112] direction by 180° is represented by the matrix

$$M = \frac{1}{3}\begin{bmatrix} -2 & 1 & 2 \\ 1 & -2 & 2 \\ 2 & 2 & 1 \end{bmatrix},$$

and one has $(hkl)M = (\overline{h}\,\overline{k}\,\overline{l})$. On the other hand, that rotation is not a crystal symmetry operation. Therefore, there are two non-equivalent orientations leading to the same pattern; if the first is represented by $O$, the second is $MO$. Let us also notice that the first-order Laue zone of this pattern is missing because of systematic extinctions; the zone consists of reflections $hkl$ such that $h + k + 2l = 1$, and this condition is satisfied only by triads $h, k, l$ containing both odd and even numbers – a recipe for extinction in f.c.c. structures.

One may argue that the above example is just a particular case in a large continuous domain of parameters and the probability of encountering ambiguities in orientation mapping is low. However, the case is not solvable for a relatively wide range of orientation parameters and microscope settings. The particular pattern shown in Fig. 1 was simulated for a voltage of 200 kV, a camera length of 180 mm, a detector size of 2.54 × 2.54 cm and a large maximal excitation error of 0.125 Å$^{-1}$. For the same voltage and the same camera length to detector size ratio, the maximal excitation error would have to be larger than 0.14 Å$^{-1}$ to obtain reflections of the higher-order Laue zone. If only voltage is changed and the other parameters are kept fixed, one needs about 108 kV to have additional weak reflections breaking the 180° ambiguity.

Moreover, the problem with indexing concerns not just a particular zone-axis pattern but also patterns nearby, because the match between an experimental pattern and a corresponding template is never perfect – neither in terms of peak intensities nor in their locations. Fast automatic indexing relies on simple kinematic approximation of intensities. On the other hand, measured intensities are influenced by noisy background; the influence is particularly

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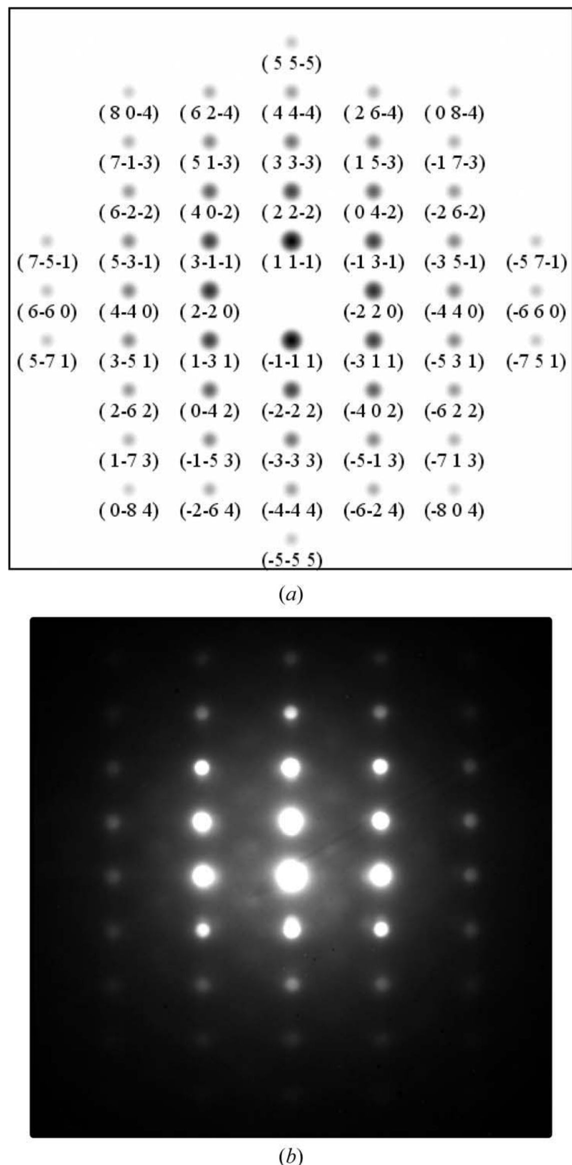


**Figure 1**
(*a*) Simulated Cu [112] diffraction pattern. A logarithm of intensities was taken and inverted gray scale is used to enhance the visibility of weak reflections. (*b*) An experimental near [112] diffraction pattern.

strong for weak reflections, which include large-angle reflections, and the latter are frequently crucial for discriminating between potential solutions. Moreover, the template easily 'misses' narrow large-angle reflections because the pattern center and camera length are known with a limited accuracy.

The reliability of solutions is measured by a properly defined (confidence) index.[1] For experimental patterns, the value of that index is influenced by numerous factors involved in pattern acquisition. To obtain an insight into the major causes of ambiguities, it is better to calculate the reliability index for simulated diffraction patterns. By computing the index as a function of direct beam direction for simulated data, one can identify crystal orientations difficult to determine in actual experiments. Example results of such a scan through all beam directions for Cu under the conditions given above are shown in Fig. 2(*a*). Besides the near ⟨112⟩ patterns, which constitute the largest fraction of those causing problems, the ⟨013⟩ and near ⟨110⟩ directions also have low reliability. The last case is an example of a minimum that is not due to the 180° ambiguity but is a result of probing the orientation space with finite steps and particular settings of the program (optimized for a set of experimental patterns); the location and depth of such minima depends on the choice of program parameters. The reliability index *versus* beam direction calculated for b.c.c. (ferrite) and hexagonal (Ti) structures is shown in Figs. 2(*b*) and 2(*c*), respectively. In the first case, the low reliability orientations are spread around the ⟨111⟩ and ⟨113⟩ directions. As in the case of the f.c.c. ⟨112⟩ and ⟨013⟩ patterns, the first-order Laue zones in b.c.c. ⟨111⟩ and ⟨113⟩ patterns are missing because of systematic extinctions. The reliability index for Ti is generally lower than that for the cubic structures, with the largest depression around the ⟨2$\bar{1}\bar{1}$5⟩ direction.

Of course, these figures are not universal. The diameter of the spread of low reliability areas depends on the experimental setup,

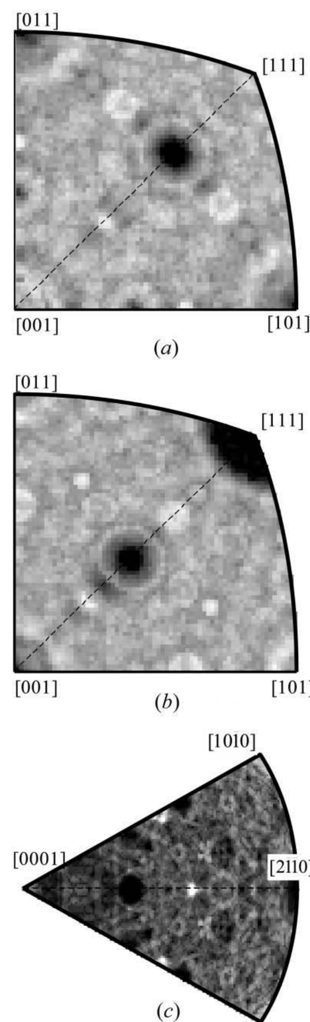


**Figure 2**
The dependence of the reliability index on the direct beam direction for simulated patterns (equal area projection). Arbitrary units are used because maximal values on the graph depend on the parameters of the indexing program, and what matters here is only the location of low values (dark areas). The patterns were simulated for Cu (*a*), ferrite (*b*) and Ti (*c*).

[1] We use a reliability index similar to that of Rauch & Dupuy (2005). It is defined as $1 - m_2/m_1$, where $m_i$ is a measure of the match between the pattern and the $i$th best template, and the orientations corresponding to $m_1$ and $m_2$ differ by at least 7°.



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including the pattern acquisition system and the indexing program. Although the program used in our computations was based on the same principles as that of Rauch & Dupuy (2005), these two implementations are likely to give slightly different results. However, the general features of such figures will be the same. For experimental patterns, the fraction of problematic cases will be larger than in Fig. 2 because of experimental errors absent in simulated patterns.

Generally, the problem can be alleviated by increasing the curvature of the Ewald sphere, *i.e.* by reducing the accelerating voltage. However, simulations show that relatively large reductions are needed, and such voltage changes may be limited by other aspects of the measurement.

Summarizing, the reliability of indexing based on electron diffraction patterns is strongly correlated to the (crystallographic) direction of the direct beam, and, with the currently used approach, it is difficult to avoid misindexing. The automatic indexing of such patterns is influenced by numerous factors but the presence of ambiguous solutions is a norm rather than an exception, and the method must be used with caution. Before proceeding with a measurement for a given material, one may check the reliability index as a function of the direct beam direction for simulated data. By repeating this exercise in various configurations, one can determine optimal microscope operating conditions, in which problematic patterns constitute the smallest possible fraction.

Finally, we would like to stress that, despite the described problem, if applied with care, automatic orientation determination using electron diffraction patterns can be an important complement of existing methods of texture analysis. Particularly promising is its application to orientation mapping of highly deformed materials for which other techniques fail.

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