Study of lattice systems separability with the use of parametric identification methods for three-dimensional crystal lattices

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Abstract. The paper deals with the problem of lattice systems overlapping, arising in structural identification of crystal lattices in three-dimensional space. The problem leads to a crucial decrease in the accuracy of structural identification and is most evident for four types of lattice systems: orthorhombic, trigonal, tetragonal and cubic. The search for separation boundaries of these systems is based on the use of three parametric identification methods (Bravais unit cell parameters, Wigner-Seitz cell volumes and distances between isosurfaces) and evaluation of several similarity measures for the estimated parameters. Computational experiments were carried out on a large set of simulated reference lattices, by which the type of the distorted lattice was identified. Research results confirmed the existence of the problem of separability for the specified lattice systems, even in the case of ideal lattices (without any distortion). However, the main result was the determination of the parameters distortion range (5\% for the edges and 3\% for the angles), within which the lattice type is identified correctly.

1. Introduction
The modern science has made a huge leap in the field of studying the matter: the possibility of obtaining high-resolution nanoscale images with the subsequent restoration of three-dimensional structure allow to study the structure of the matter itself. In this area, particular attention has always been paid to crystals – the substances with an ordered structure. The relation between the geometrical structure and physicochemical properties is observed most clearly [1,2] in a crystal.

In order to describe the structure of a crystal, a mathematical abstraction was proposed – a crystal lattice, which is a 3-D periodic structure obtained by translation of a single unit cell (minimum building block) [3,4]. A unit cell can be defined by three vectors or by six parameters: the lengths of three edges and the measures of three angles between them (figure 1).

The most well-known way to describe a crystal lattice is the method proposed by Auguste Bravais [5]. There are only 14 three-dimensional Bravais lattices which can form any crystal lattice. The Bravais lattices are divided into 7 crystal systems, which can be described by three translation vectors (figure 2).
However, there still remains the key problem of determining the type of crystal lattice according to the data of X-ray diffraction analysis. The main way to solve it is to compare the parameters estimated for the lattice under study with the reference parameters [6]. The following two circumstances make this problem particularly complicated: the ambiguity of the unit cell choice [7] and the overlapping of crystal lattice classes [8]. While the ambiguity of choice can be eliminated by introducing the criterion of lattice nesting, the second circumstance is much more complicated.

The previous studies on the structural identification of crystal lattices [9, 10] showed that the greatest difficulty arises when identifying the lattices of the following four crystal systems: orthorhombic, trigonal, tetragonal, and cubic. The reason for this is the overlapping of classes of crystal lattices, which have not been managed even with the help of neural networks [9]. In this regard, the purpose of this work was to determine the boundaries of separability of these four types of crystal systems using the methods of crystal lattices parametric identification.

### Figure 2. Types of Bravais lattices and their division into crystal systems.

2. The methods of 3-D crystal lattices parametric identification

The basis of the planned study is the selection of a number of the lattice individual geometrical parameters and their subsequent comparison with the reference ones. For this purpose, we chose the following three developed methods of parametric identification with the normalized similarity measures of the evaluated parameters.

1. The method of parametric identification based on the evaluation of Bravais unit cells. This method estimates the six basic parameters of the Bravais unit cell — the lengths of three edges and the measures of three angles between the edges [8, 10, 11].

The measure of edges similarity:
\[ \| \mathbf{f} - \mathbf{r} \| = 1 - \frac{\sqrt{(l_{11} - l_{11})^2 + (l_{12} - l_{22})^2 + (l_{13} - l_{33})^2}}{\max \left\{ \sqrt{(l_{11})^2 + (l_{12})^2 + (l_{13})^2}, \sqrt{(l_{22})^2 + (l_{22})^2 + (l_{23})^2} \right\}}. \]

The measure of angles similarity:

\[ \| \mathbf{\alpha}_1 - \mathbf{\alpha}_1 \| = 1 - \max \left\{ \sin (|\alpha_{11} - \alpha_{21}|), \sin (|\alpha_{12} - \alpha_{22}|), \sin (|\alpha_{13} - \alpha_{23}|) \right\}. \]

The measure of G6 coordinates similarity \([11, 12]:\)

\[ \| \mathbf{g}_1 - \mathbf{g}_2 \| = 1 - \max_{l \in \mathbb{C}^6} \left\{ \left| g_{1l} - g_{2l} \right| \right\}. \]

2. The method of parametric identification based on the evaluation of Wigner-Seitz cells. This method estimates the volume of a Wigner-Seitz cell \(V\) \([8]\).
The measure of similarity of volumes:

\[ \| V_1 - V_2 \| = 1 - \frac{\| V_1 - V_2 \|}{\max \{V_1, V_2\}}. \]

3. The method of parametric identification based on the evaluation of distances between the isosurfaces \([13]\). The essence of the method is the development of a set of isosurfaces covering the unit cell, and calculation of three distances between them: the average root-mean-square distance \((\text{avg } d_{RMS})\); the average and the maximum Hausdorff distances \((\text{avg } d_H \text{ and } \text{max } d_H)\).

The measure of distances similarity:

\[ \| d_1 - d_2 \| = 1 - \frac{\| d_1 - d_2 \|}{\max \{d_1, d_2\}}. \]

3. Scheme of the computational experiment

The purpose of the study presented in this paper was to determine the limits of variation of the unit cell parameter (edge or angle), at which this cell ceases to belong to the original type of the crystal system and becomes similar to another cell type.

A cubic lattice with the following parameters was studied:

- edge lengths – 5 Å;
- angles – 90°;
- lattice size – 2 translations in each direction (27 nodes).

The choice of this unit cell type is determined by the possibility to change any one or two of its parameters independently (edge or angle).

The computational experiments aimed at the study of separability of various types of crystal systems using structural identification methods were carried out according to the following scheme:

1. 1000 random lattices are simulated for each type of unit cell with the following parameters:
   - edge lengths – in the range from 4 to 6 Å;
   - angles \(\alpha_1, \alpha_2\) – in the range from 85° to 95°, \(\alpha_3\) – in the range from 85° to 95°;
   - lattice size – 2 translations in each direction (27 nodes).

2. The parameters of each lattice are evaluated using the three identification methods chosen: edge lengths, angle measures and cell volume, coordinates in the 6-dimensional G6 space, distances between the sets of isosurfaces.

3. One chosen parameter is changed successively and the resulting lattice is compared with all the simulated lattices, the number of the matching lattices of each crystal system is calculated. Similarity criterion: all the similarity measures described take a value of at least 0.95.

Due to the random nature of the experiment, the average results of 20 repeated experiments were selected as the final values of the average parameter identification errors to ensure the accuracy of the parameters calculation of at least 0.95 and reliability at the level of 0.99.

It should also be noted that the design of the experiment is computation-intensive, for this reason, previously developed efficient parallel implementations of parametric identification methods were
used [14]. The parallelism model integrated in the methods allows to achieve the acceleration close to linear even for the lattices with a small number of nodes. All computations were made on the “Sergey Korolev” cluster: 16 nodes, each based on two four-core Intel Xeon X5560 processors.

4. Study of the dependence of structural identification on the distortion of one parameter
First of all, the dependence of the structural identification of crystal lattices on the distortion of one edge of the unit cell was analyzed. The edge $l_1$ was changed uniformly with a step of 0.02 Å in the range from 4 to 6 Å.

Let us present the results obtained in the form of a diagram showing the dependence of structural identification on the distortion of a unit cell edge (figure 3).

![Figure 3. Dependence of structural identification on the distortion of a unit cell edge.](image)

The above diagram shows that the largest percentage of the matching lattices are the cubic-type lattices like the one under study. The tetragonal and orthorhombic cells differ from the cubic cell in the lengths of one and two edges respectively, so over the entire range of variation of the $l_1$ edge, the matching of the lattice under study with those of the above two types can be observed.

Special attention should be paid to the fact that the matching with trigonal-type lattices (blue graph) occur within the same boundaries as the matching with cubic-type structures (red graph). This result is explained by the fact that a trigonal cell, like the cubic one, has equal edges, therefore a distortion of one of the edges leads to the same distortion in the similarity measures of both types and, consequently, to the same deviation from the original type of the crystal system.

A similar study was carried out to determine the dependence of crystal lattice structural identification on the distortion of one angle of a unit cell. The angle $\alpha_1$ changed uniformly with a step of 0.1° in the range from 85 to 95°.

According to the data obtained, a diagram was developed showing the dependence of structural identification of the lattice under study on the distortion of a unit cell angle (figure 4).

The presented diagram shows that the largest percentage of the matching lattices are the cubic-type lattices like the one under study. The tetragonal and orthorhombic cells have the same angles as the cubic cell, so the boundaries of matching with the cells of the three types are equal. It should be noted, that the boundaries of matching with trigonal cells are wider as compared to other types. This result is explained by the fact that the angles of a trigonal cell are not 90°, therefore the matching may occur at the boundaries of the used range of angles, but non-angle-related measures of similarity are more important in these areas.

It should be noted that the resulting diagram is symmetric in 90°. This feature proves that the identification methods proposed can be equally applied to the cells with both acute and obtuse angles.
5. Conclusion
The studies conducted in this paper have confirmed the existence of the separability problem for the four types of lattice systems: cubic, trigonal, tetragonal and orthorhombic. As it was shown, even in the ideal case (without any distortion of the lattice parameters, which is absolutely impossible in practice), there are matching with other types of lattices. In other words, two lattices physically belonging to different crystal systems, are indistinguishable in practice in case of small distortions of coordinates of the nodes.

On the other hand, based on the results obtained, it can be concluded that if the edges deviate by no more than 5% and the angles deviate by no more than 3%, the lattice type will still be identified correctly. This result is achieved through the use of distortion resistant identification methods and a large base of reference lattices, which leads to a greater number of matches with the lattices of the initial type.

However, it should be noted that the simple increase in the size of the database of reference lattices covering the whole space of the Bravais lattices parameters uniformly will produce an adverse effect on the accuracy of structural identification. The specified “corridor” for the edges and angles shall be observed in order to separate the lattices of various types of lattice systems.

6. References
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