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Inventorying quasicrystal atomic surfaces by watershed algorithm

Pavel Kalugin, André Katz

► **To cite this version:**

Pavel Kalugin, André Katz. Inventorying quasicrystal atomic surfaces by watershed algorithm. Open space between aperiodic order and strong electronic correlations, Jun 2017, Annecy, France. Unpublished, 2017, 10.13140/RG.2.2.26140.85127 . hal-04215070

HAL Id: hal-04215070

<https://hal.science/hal-04215070>

Submitted on 26 Sep 2023

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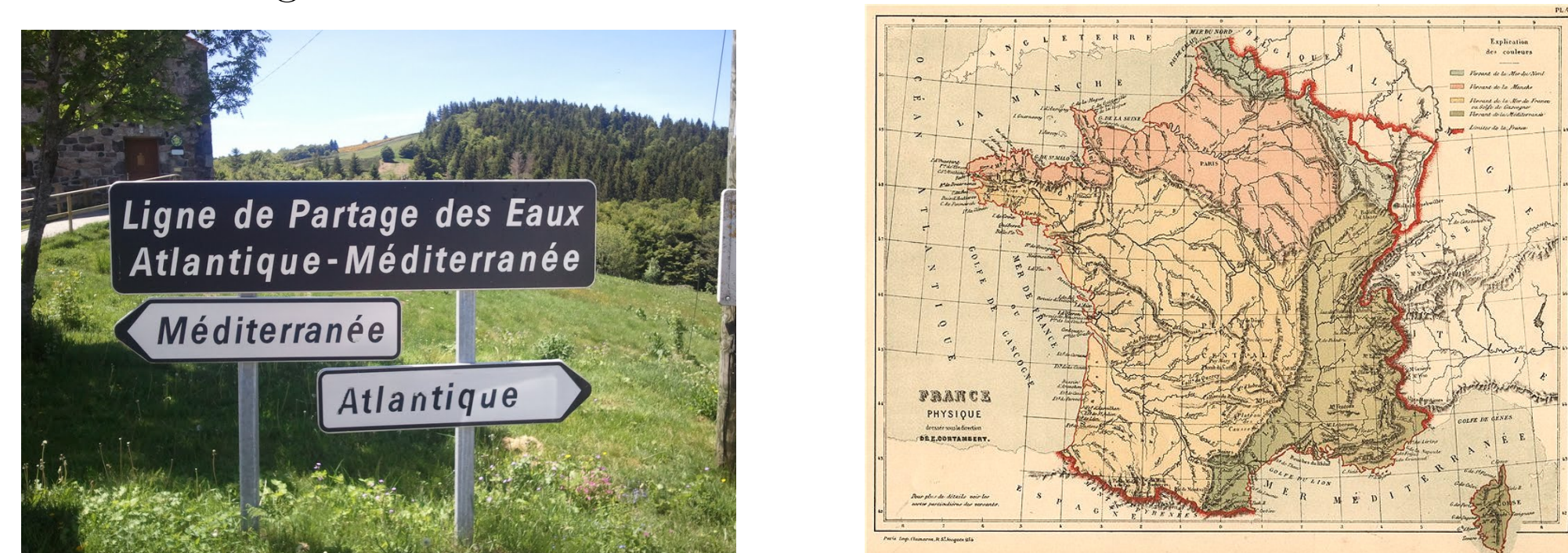
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Motivation

The traditional way of assessing crystal structures out of phased diffraction data consists in visual examination of 2D cuts through density maps. However, for icosahedral quasicrystals such maps are six-dimensional, making direct human examination unreliable. We have developed an automated procedure for assessing location of atomic surfaces, to make sure that all matter is taken into account. Our approach consists in generalization of classical watershed segmentation algorithm to 6D space.

Watershed segmentation

The idea of the watershed algorithm is inspired by the geological phenomenon of the drainage divide:



In application to the digital image processing the gray levels are assimilated to an altitude of a topographical surface. The watershed algorithm separates the image into catchment basins divided by crest lines:

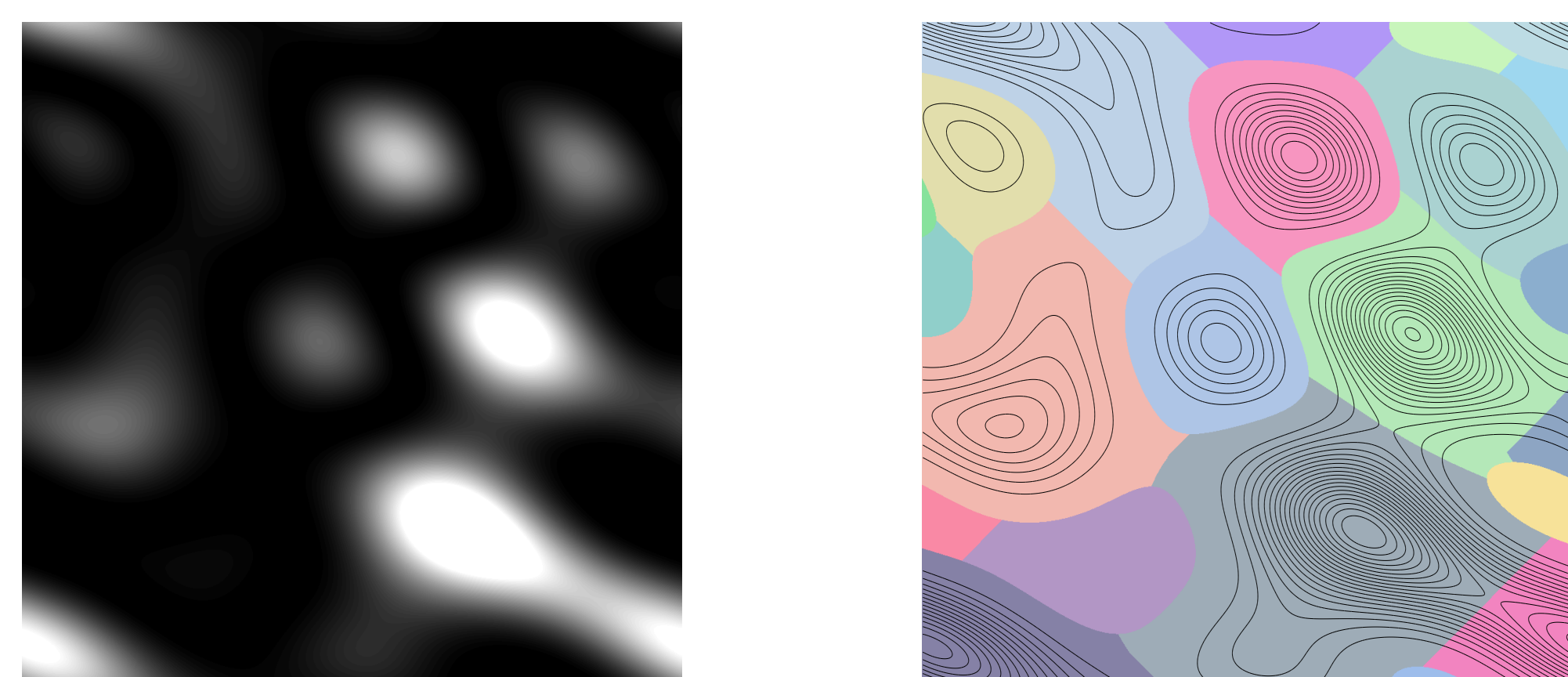


Figure: Left: gray level representation of a scalar test function. Right: watershed basins (marked by arbitrary colors) with level lines superimposed.

There exist many versions of watershed algorithm, most of which are variants of the Priority Flood method introduced in [1]. Watershed segmentation can be applied to any regular scalar function on the plane, with the catchment basins constructed around either the local minima or maxima. Practical application of the algorithm requires sampling of the function on a lattice and definition of the nearest neighbors connectivity graph.

Segmentation in six dimensions

The Priority Flood method of [1] admits a straightforward generalization to any spatial dimension and thus could be used to separate atomic surfaces of icosahedral quasicrystals by segmentation of 6D density maps. However, the standard algorithm is not applicable *as cast*, because of the following reasons:

- **Curse of Dimensionality.** In six dimensions the two-fold improvement of the spatial resolution comes at the cost of the 64-fold increase of the volume of data.
- **Oversegmentation.** This phenomenon is an artifact of the sampling procedure, which may transform a single local minimum into a cluster of minima, each yielding an independent basin. Oversegmentation is aggravated by strong anisotropy of the data, which is the case for the 6D density maps for quasicrystals.

Oversegmentation can be alleviated by the choice of the nearest neighbors graph with higher connectivity, while the residual over-split basins can be joined together by the so-called “waterfall” algorithm [2]. However, these approaches alone do not suffice to make watershed practicable for the real 6D density data. We have developed a construction of the sampling lattice which respects the anisotropy of the data and makes watershed segmentation feasible for quasicrystals.

Data preparation

We tested our algorithm with the diffraction data taken from the supplemental materials to [3]. Only the peaks with intensity exceeding the uncertainty were kept, leaving 5078 independent reflections. Normalized structure factors were obtained with the atomic form factors by Waasmaier and Kirfel averaged with respect to the composition and assuming the B-factor equal to that of metallic Cd under the same conditions (1.8\AA^2).

The phasing was performed by a custom version of charge flipping algorithm. To suppress oscillations of the charge density, the normalized structure factors were multiplied before phasing by the window function

$$w(k) = \frac{(k_{\max} - |k|)^2(2k_{\max} + |k|)}{2k_{\max}^3},$$

where $k_{\max} = \max(k)$. This filtering was applied for both parallel and perpendicular components of the wave vector. However, the watershed segmentation was performed with unfiltered data to allow for better separation of atomic surfaces.

Handling anisotropy

The anisotropy of the density map can be characterized by the covariance matrix:

$$Q_{ij} = \sum_n I_n k_i^{(n)} k_j^{(n)},$$

where I_n stands for the intensity of the n -th Bragg peak and $k_i^{(n)}$ are the components of the corresponding 6D reciprocal lattice vector. To compensate for the effect of anisotropy we start with a dense packing of spheres in \mathbb{R}^6 equipped with the metric Q_{ij} instead of the Euclidean one. This packing needs to be slightly deformed to respect the periodicity of the 6D density, in such a way that the sampling lattice becomes a super-lattice of \mathbb{Z}^6 .

In practice, we start with the Euclidean lattice D_6 (with 60 nearest neighbors for each point) and construct out of it a set of 119347420 sampling points. The Priority Flood step of segmentation produced 20938 basins. After the waterfall step of the algorithm, only 58 basins were formed by fusion of over-segmented fragments (see details below).

Result for i-Cd_{5.7}Yb: five atomic surfaces

The 58 watershed basins are organized in five orbits of the icosahedral symmetry group. We label three of these orbits by symbols V, B and E, following the notations of [3]. Two other orbits, which we label as S2 and S3, were not considered in [3] as separate atomic surfaces but rather as parts of the surfaces B and V respectively. The figures represent two-dimensional cuts of the watershed basins (identified by the color code below) along various symmetry axes. The (filtered) density map is represented by its level lines, superimposed on the plots.

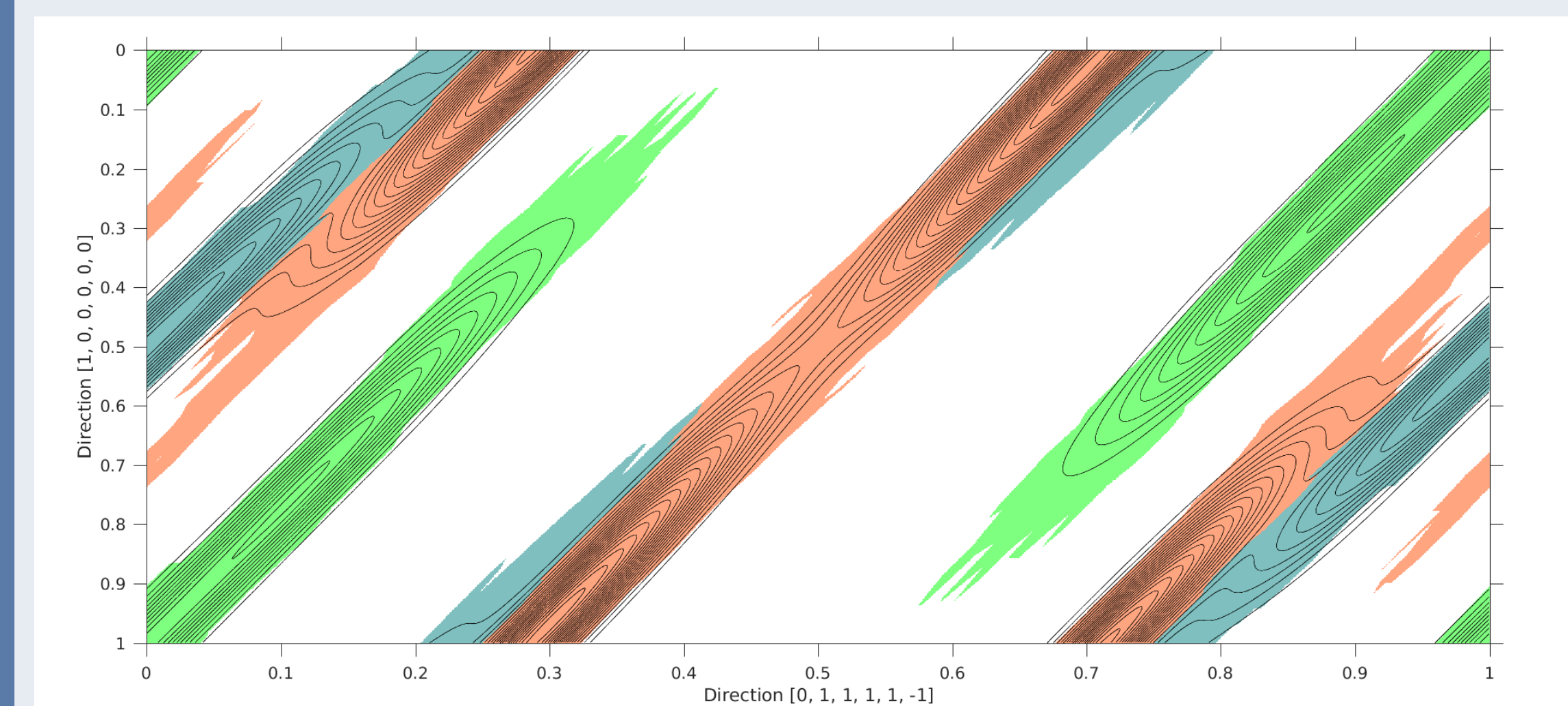


Figure: Five-fold axis.

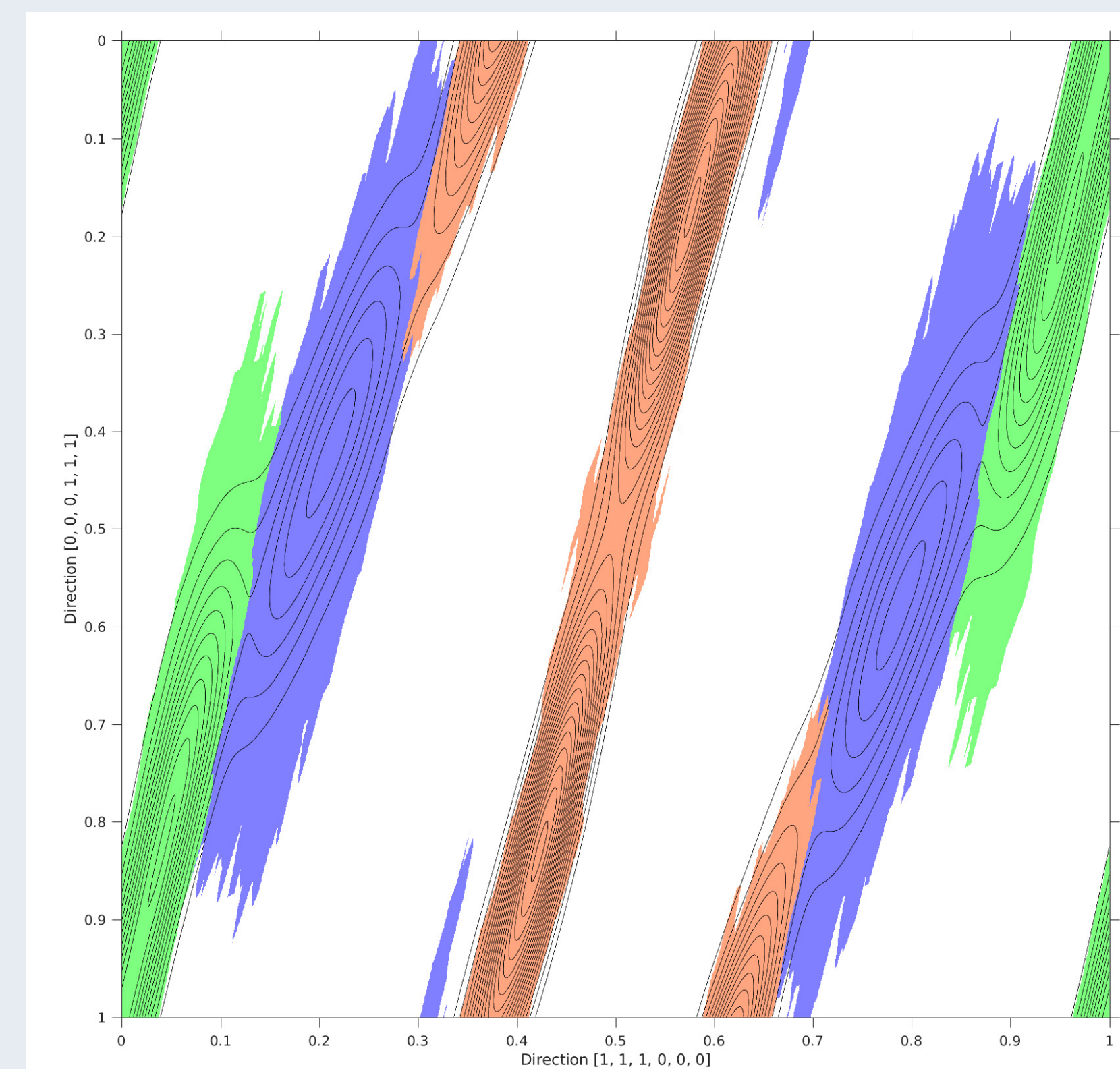


Figure: Three-fold axis.

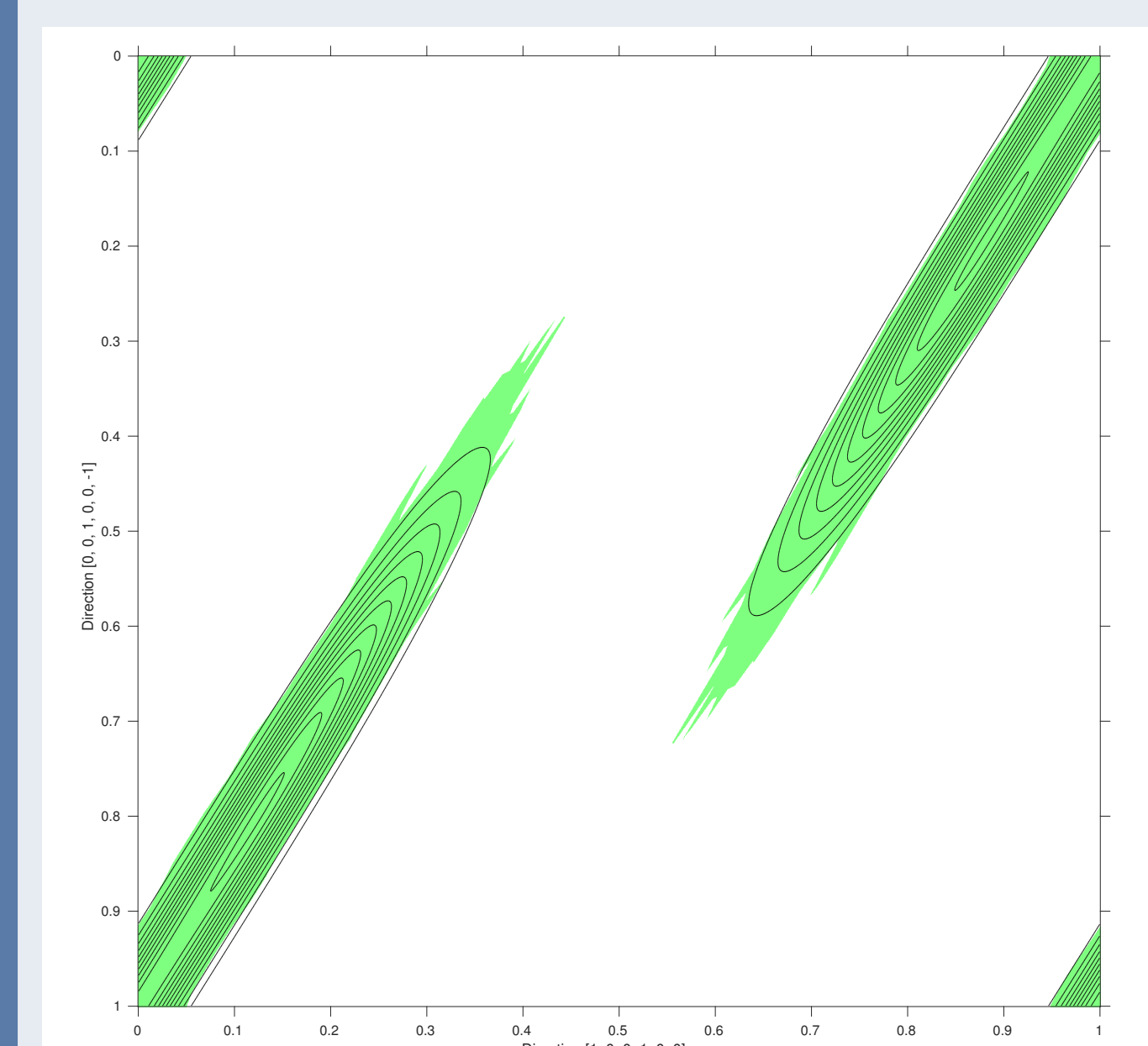


Figure: Two-fold axis going through $[0, 0, 0, 0, 0, 0]$.

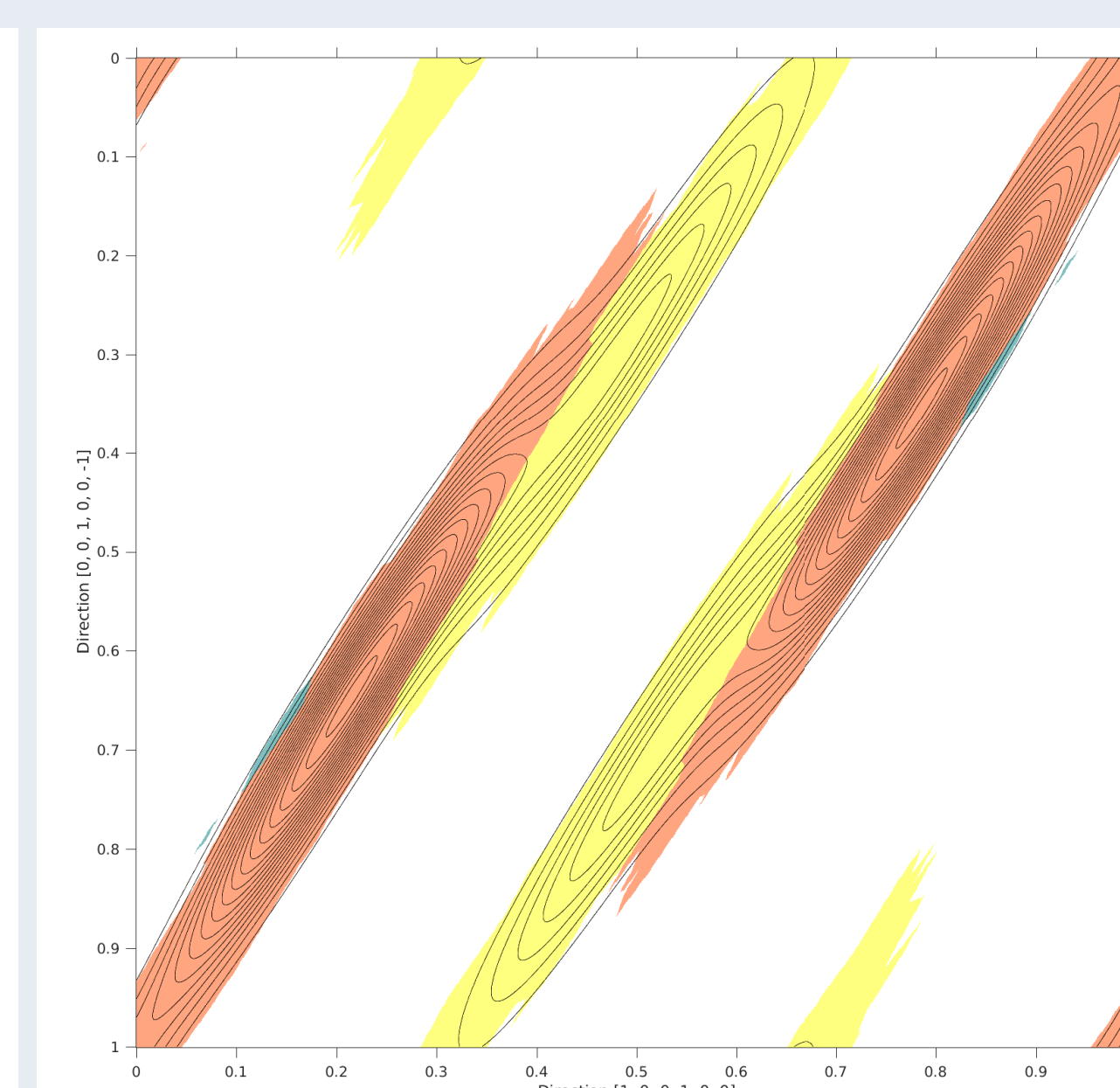


Figure: Two-fold axis going through $[\frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}]$.

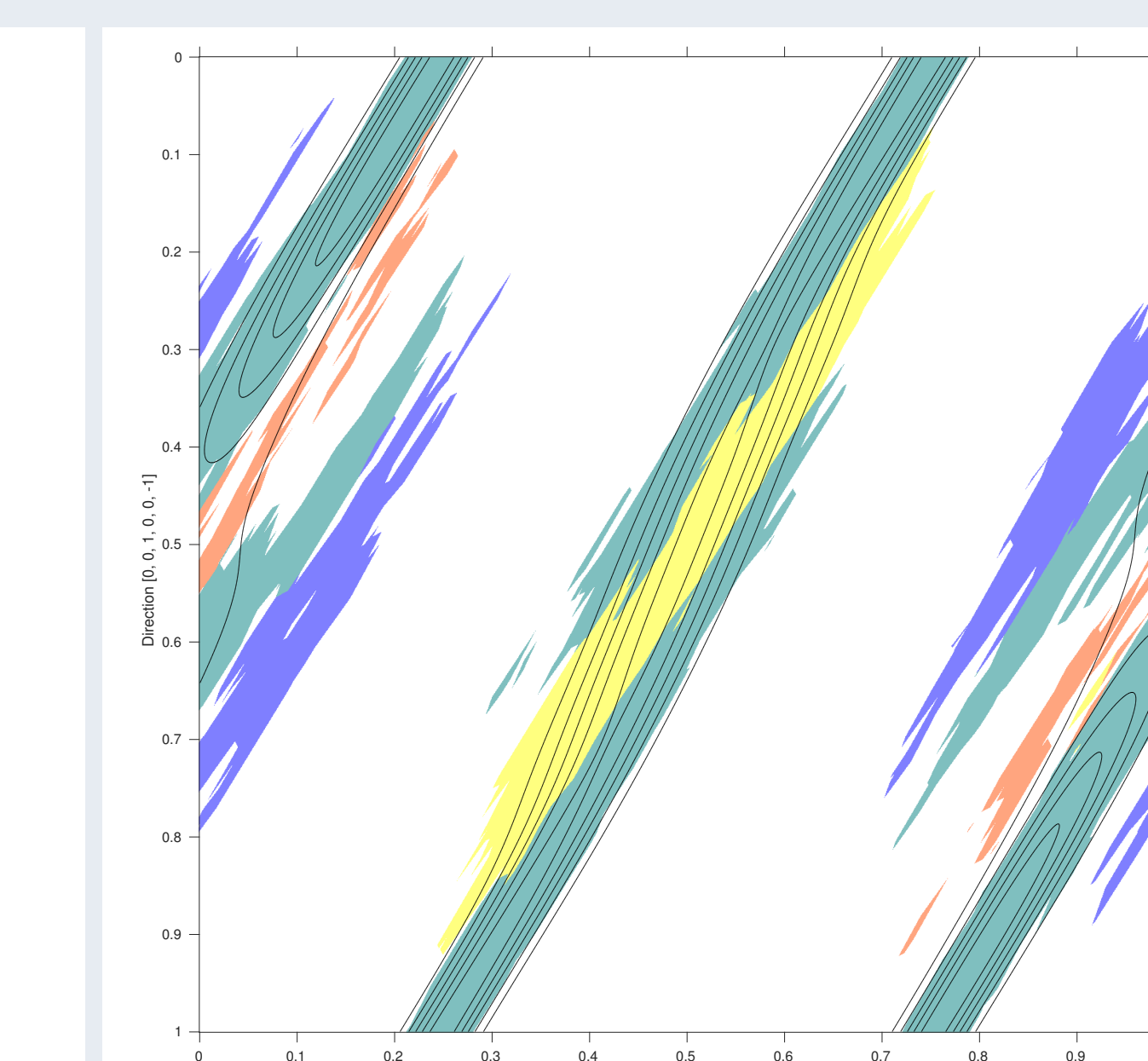


Figure: Two-fold axis going through the point of the maximal density of the atomic surface S2. Note the surprising proximity of the S2 and E surfaces.

- V: single atomic surface with full icosahedral symmetry, centered at $[0, 0, 0, 0, 0, 0]$.
- B: idem, centered at $[\frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}]$
- E: an orbit of 6 atomic surfaces located at $[\frac{1}{2}, 0, 0, 0, 0, 0]$ and equivalent points.
- S3: an orbit of 20 atomic surfaces located at a generic position on the three fold symmetry axis.
- S2: an orbit of 30 atomic surfaces located at a generic position on the two fold symmetry axis going through $[\frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}]$.

Sequel: diffractive power

The density maps are commonly used to find positions of the atomic surfaces, while the amplitude of the density peaks is mostly overlooked. Indeed, these amplitudes may be strongly influenced by local Debye-Waller factor or by insufficient resolution in the perpendicular space. However, neither of these phenomena affects the integral of the density over each individual atomic surface, that is the total diffractive power of the latter.

We provide, for the first time, an efficient way to assess this quantity directly from the experimental data.

The sampling lattice itself provides a tool to sum the density over the watershed basins. However, the result of such an integration is biased because of the unknown constant term (the average density). This is why one has to keep track of the volume of the basins as well:

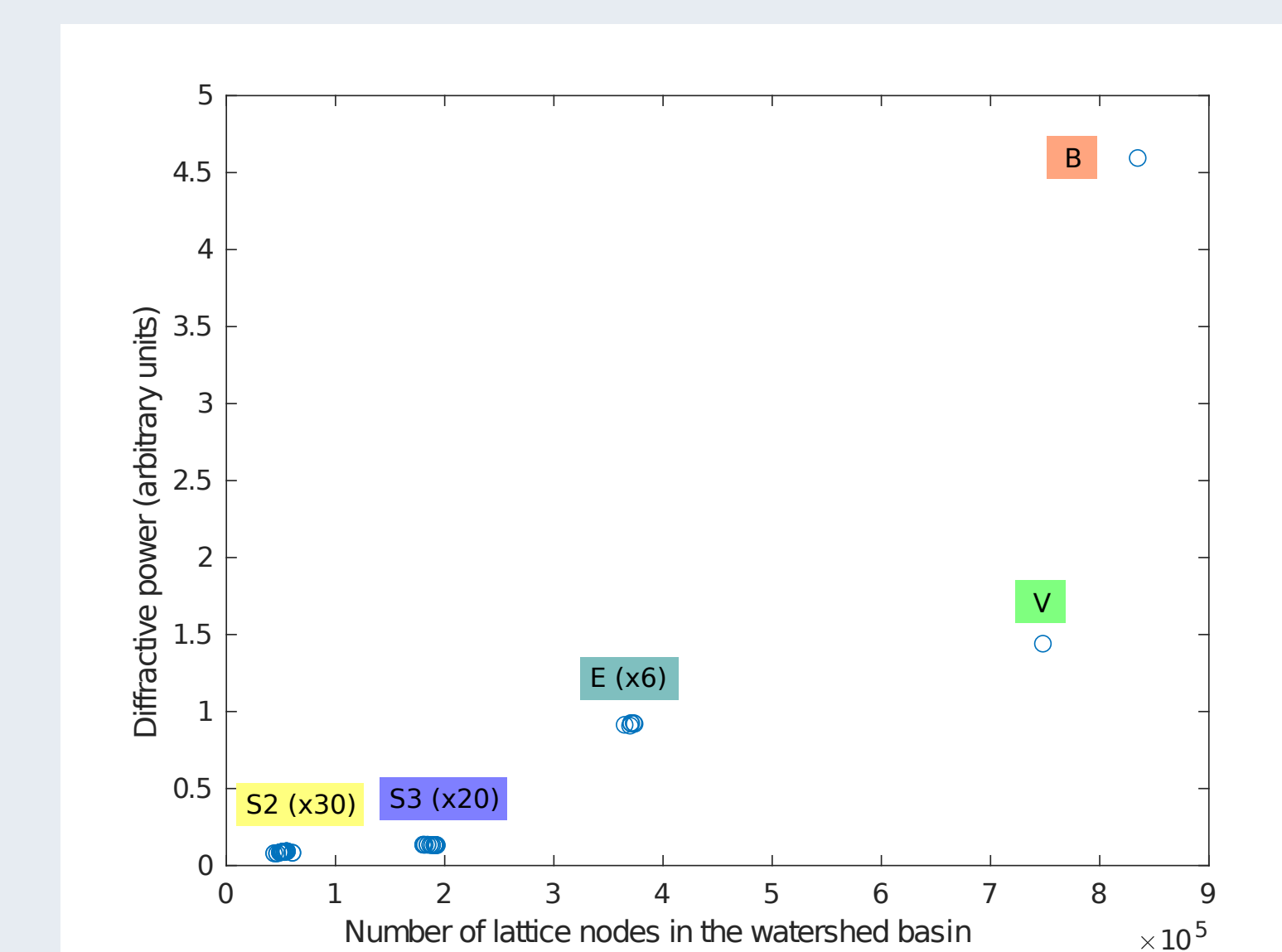


Figure: Electron density integrated over watershed basins versus their volumes (no correction for the constant term made).

Since the watershed basins of the atomic surfaces occupy less than 8% of the volume of the unit cell, one can use the average density in the rest of the volume to estimate the constant term and apply the corresponding correction. The results are summarized in the following table:

Atomic surface	Diffractive power
1 × B	0.264
1 × V	0.087
6 × E	0.326
20 × S3	0.174
30 × S2	0.149

Table: Distribution of the diffractive power (totaling 1) between orbits of atomic surfaces.

These results may be used to estimate the perpendicular volume of the atomic surface prior to any structure modeling. They also provide insight for the chemical composition of individual atomic surfaces. For instance, these data imply a mixed composition of the atomic surface B, since the full diffractive power of Yb in $\text{Cd}_{5.7}\text{Yb}$ amounts only to 0.204 of the total.

References

- [1] The morphological approach to segmentation: the watershed transformation. S.Beucher and F.Meyer, *Optical Engineering*, **34**, (1992), p. 433.
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- [3] Atomic structure of the binary icosahedral Yb-Cd quasicrystal. Takakura, H., Gómez, C. P., Yamamoto, A., De Boissieu, M., and Tsai, A. P., *Nature materials*, **6**(1), (2007), p. 58

Contact Information

- Email: kalugin@lps.u-psud.fr
- Phone: +33-1-69-15-69-39