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Periodic Sphere Packings and the Wulff-Shape

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Abstract. The asymptotic shape of large densest sphere packings in a lattice $L \subset E^d (d \geq 2)$, measured by parametric density, is a polytope, the so-called Wulff-shape of ideal crystals in crystallography. This was shown in [15] and [16] by density deviation derived from parametric density. Our main goal is a generalization of this approach from lattice packings to periodic packings of spheres. It turns out that the asymptotic shape of large densest periodic packings is a generalized Wulff-shape which is more complicate than in the lattice case. As an application we determine this Wulff-shape for a type of crystals with two different atoms (NaCl-type) and for a non-lattice crystal (diamond) and for a combination of both (ZnS-type). In all cases the result coincides for suitable parameter with the natural shapes. In particular the asymptotic shape in the case of the ZnS-type is a tetrahedron, i.e. not centrally symmetric. Although the intention of these generalizations is application to crystallography, the results can also be considered as contributions to finite and infinite packings of spheres.

1. Introduction

Let B^d denote the unit ball in Euclidean d-space $E^d (d \geq 2)$. Let V denote the volume and $V(B^d) = \kappa_d$. For $x, y \in E^d$ let xy denote their scalar product, so that $xx = \|x\|^2$. For $X \subset E^d$ let $\operatorname{lin}(X)$, $\operatorname{aff}(X)$, $\operatorname{pos}(X)$ and $\operatorname{conv}(X)$ be its linear, affine, positive and convex hull, respectively. In the following let $\mathcal{L}^d$ denote the set of lattices $L \subset E^d$ with $\det L \neq 0$. For $L \in \mathcal{L}^d$ let $\lambda_i(L) = \lambda_i(B^d, L), i = 1, \ldots, d$, and $\mu(L) = \mu(B^d, L)$ be its successive minima and its inhomogenous minimum (or covering radius), respectively (see e.g. [10]). In particular $\lambda_1(L)$ is the length of the shortest nonzero vector in L. Let $\mathcal{P}(L)$ be the set of polytopes P with nonempty interior and facets orthogonal to suitable vectors of the polar lattice L^*. In particular, $\mathcal{P}(L)$ contains the set of L-polytopes, i.e. convex hulls of finite subsets of L.

If $C_n = \{c_1, \dots, c_n\} \subset E^d$ is a discrete set such that $C_n + B^d$ is a packing, then its parametric density is defined (see [14]) by

$$\delta_\varrho(B^d, C_n) = n\kappa_d / V(\operatorname{conv}(C_n + \varrho B^d)), \tag{1}$$

where $\varrho > 0$ is a parameter. For a convex body K and a packing lattice $L \in \mathcal{L}^d$ for K we use $\delta_\varrho(B^d, K) = \delta_\varrho(B^d, L, K)$ instead of $\delta_\varrho(B^d, K \cap L)$. In the case of lattice packings for any sequence $P_n \in \mathcal{P}(L)$, whose inradii tend to infinity, we have (cf. e.g. [10], p.219)

$$\limsup_{n\to\infty} \delta_\varrho(B^d, P_n) = \kappa_d / \det L =: \delta_L, \tag{2}$$

where δ_L is the lattice packing density for B^d and L. So as a refinement the density deviation for $P \in \mathcal{P}(L)$ has been defined ([15]) as

$$\Delta_L(P, \varrho) = \lim_{\lambda\to\infty} (V(\lambda P))^{1/d} \{\delta_L - \delta_\varrho(B^d, \lambda P)\}. \tag{3}$$

In [15] it was proved that for $P, Q \in \mathcal{P}(L)$ with $\Delta_L(P, \varrho) < \Delta_L(Q, \varrho)$, P is, in a natural sense, asymptotically ϱ-denser than Q, and the minimum of $\Delta_L(P, \varrho)$ is attained for the Wulff-shape

$$W(L, \varrho) = \left\{ x \in E^d \ : \ ux \le \varrho \|u\| - \frac{1}{2}, \ u \in L^* \text{ primitive} \right\}. \tag{4}$$

$W(L, \varrho)$ is a polytope and it depends only on L and ϱ. Only ϱ with

$$\varrho > \varrho_L = \max\{(2\|u\|)^{-1} : u \in L^* \setminus \{0\}\}$$

are considered. This guarantees that $W(L, \varrho)$ is nonempty and no $(d-1)$-dimensional P is ϱ-denser than all $P \in \mathcal{P}(L)$.

In Section 2 we give an alternative proof that $W(L, \varrho)$ is a polytope, without using diophantine approximation and give bounds for the number of facets depending on ϱ.

It is remarkable that the principle of minimizing the volume (maximizing the density) is equivalent to the principle of minimizing the free surface energy, which is known as the Gibbs-Curie-Wulff principle (cf. e.g. [11], [5]). Another approach is in [1], where the asymptotic shape of the densest packing configurations $C_n \subset L$, i.e. with maximal $\delta_\varrho(B^d, C_n)$ for given n, is considered.

Our main goal is a generalization of Wills' approach from lattice packings to periodic packings of spheres. Only few crystals are 1-atomic lattice packings (e.g. metals and rare gases). So a generalization to periodic packings of spheres with different radii is reasonable. For this we need some more notations.

Let $K_1, \dots, K_n$ be convex bodies with volumes $v_1, \dots, v_n$ and $v = v_1 + \dots + v_n$. Further let $L \in \mathcal{L}^d$ and for $t_1, \dots, t_n \in E^d$ let $L_j = L + t_j$, $j = 1, \dots, n$ (we will always assume that $t_1 = 0$). If $(L_1 + K_1) \cup \dots \cup (L_n + K_n)$ is a packing then $M = L_1 \cup \dots \cup L_n$ is a periodic packing for $K_1, \ldots, K_n$ (cf. [12]). In the following we use M for the point set together with the K_i and the v_i. All ideal crystals can be presented in this way and in general M is not a lattice (although chemists speak of lattices). In this application $K_1, \ldots, K_n$ are spheres (atoms) of different size.

For a convex body K we define $L_j(K) = \operatorname{card}(K \cap L_j)$ and $M(K) = \operatorname{card}(K \cap M)$ analogously to the lattice point enumerator $L(K) = G(K, L) = \operatorname{card}(K \cap L)$. Further let

$$\delta_\varrho(M, K) = \delta_\varrho(B^d, M, K) = \frac{\sum_{j=1}^n v_j L_j(K)}{V(K + \varrho B^d)} \tag{5}$$

be the parametric density with respect to M and ϱ. If K is the convex hull of M-points, then (5) is a generalization of (1). Further the definition is natural, if $K_1, \ldots, K_n$ are balls with different radii. In general the K_i have different shapes and the question is how to choose a gauge body C instead of B^d in the denominator. For the choice of C one can take chemical and physical aspects into consideration, but in this paper we choose $C = B^d$ also for the general case. Note that the density in (5) may be greater than 1 for $\varrho = 1$. To avoid this one can use $C = rB^d$ with a suitable $r > 0$.

The density δ_M of an infinite periodic packing is (cf. [12], p.25)

$$\delta_M = \frac{v}{\det L}. \tag{6}$$

To generalize (3), it is not sufficient to consider only dilatates to a given $P \in \mathcal{P}(L)$ because there might be many points of M close to a facet and it is more efficient to include them. So we choose a sequence from the set of polytopes $\mathcal{P}(P) \subset \mathcal{P}(L)$, which have the same outer normals as P. Now we introduce the density deviation for periodic packings.

Definition 1. *For $P \in \mathcal{P}(L)$ let*

$$\Delta_M(P, \varrho) = \inf \liminf_{\lambda \to \infty} (V(\lambda R_\lambda))^{1/d} \{\delta_M - \delta_\varrho(M, \lambda R_\lambda)\},$$

where the infimum is taken over all sequences $\{R_\lambda\}$ with $R_\lambda \in \mathcal{P}(P)$ and $R_\lambda \to P$.

To justify the definition of the density deviation we will prove in Section 3 that if $\Delta_M(P, \varrho) < \Delta_M(Q, \varrho)$ for $P, Q \in \mathcal{P}(L)$, then P is asymptotically ϱ-denser than $Q \in \mathcal{P}(L)$ in the following natural sense.

Definition 2. *Let $P, Q \in \mathcal{P}(L)$. Then P is called asymptotically ϱ-denser than Q if there is a sequence $R_\lambda \in \mathcal{P}(P)$ with $R_\lambda \to P$ such that for each sequence $S_\nu \in \mathcal{P}(Q)$ with $S_\nu \to Q$ and each $\nu \in \mathbb{N}$ a $\lambda(\nu)$ can be chosen such that for $\nu \geq \nu_0$*

$$M(\lambda(\nu) R_{\lambda(\nu)}) \leq M(\nu S_\nu) \leq M((\lambda(\nu)+1) R_{\lambda(\nu)+1})$$

and

$$\min\left\{\delta_\varrho(M, \lambda(\nu) R_{\lambda(\nu)}), \delta_\varrho(M, (\lambda(\nu)+1) R_{\lambda(\nu)+1})\right\} > \delta_\varrho(M, \nu S_\nu).$$

For $x \in \mathbb{R}$ let $\{x\} = x - [x]$ be the fractional part of x. For primitive $u \in L^*$ and $j = 1, \ldots, n$ we define

$$\epsilon_j(u) = \{u \cdot t_j\}.$$

Then $\epsilon_j(u) \in [0, 1)$ and $\epsilon_j(u) \det L / \Delta(u)$ represents the distance of the hyperplane containing points of L_j between $\{x : ux = 0\}$ and $\{x : ux = 1\}$ to the origin, where $\Delta(u) = \det L \|u\|$ is the determinant of the $(d-1)$-sublattice of L orthogonal to u. For non-primitive $u \in L^*$ let $\epsilon(u) = \epsilon(u')$, where $u' = u/m$ primitive. Note that $\epsilon_j(-u) = 1 - \epsilon_j(u)$, for $\epsilon_j(u) \neq 0$.

Let $P \in \mathcal{P}(L)$ such that the outer normals u_i, $i = 1, \ldots, k$, of its facets F_i are primitive vectors of L^*. Let $f_1, \ldots, f_k$ denote their $(d-1)$-dimensional volumes and $\Delta_1, \ldots, \Delta_k$ the $(d-1)$-dimensional determinants of the corresponding sublattices of L. Note that $\Delta_i = \det L \|u_i\|$. For fixed u_i each L_m, $m = 1, \ldots, n$, determines a family $\mathcal{H}_m(u_i)$ of parallel hyperplanes orthogonal to u_i. For the approximating sequence R_λ in Definition 1 we have to choose for each u_i the position of the corresponding supporting hyperplanes. We will prove that R_λ leads to an optimal density, if these hyperplanes belong to one certain family $\mathcal{H}_m(u_i)$. The following definition is motivated by the choice of this optimal family.

Definition 3. *For $u \in L^*$ and $m = 1, \ldots, n$ let*

$$\tau_m(u) = \frac{1}{2} - \sum_{j=1}^{n} \frac{v_j}{v} \cdot \{\epsilon_m(u) - \epsilon_j(u)\} \tag{7}$$

and

$$\tau(u) = \max_{1 \leq m \leq n} \tau_m(u). \tag{8}$$

We call $\tau(u)/\|u\|$ the optimal facet density in direction of u.

The geometric meaning of $\tau(u)$ will be clear in the proof of the main result in Section 3

$$\Delta_M(P, \varrho) = \delta_M V(P)^{1/d-1} \sum_{i=1}^{k} f_i \left(\varrho - \frac{\tau(u_i)}{\|u_i\|} \right), \tag{9}$$

and there is a $P \in \mathcal{P}(L)$ with minimal $\Delta_M(P, \varrho)$, which is given by the following

Definition 4. *Let*

$$W_M = W(M, \varrho) = \{x : ux \leq \varrho \|u\| - \tau(u),\ u \in L^* \text{ primitive}\} \tag{10}$$

be the generalized Wulff-shape.

For $n = 1$ we have $\tau(u) = 1/2$ and hence W_M is a generalization of $W(L, \varrho)$ in (4). We only consider such ϱ with

$$\varrho > \varrho_M := \max \left\{ \frac{\tau(u)}{\|u\|} : u \in L^* \backslash \{0\} \right\}. \tag{11}$$

This guarantees that W_M is nonempty and no $(d-1)$-dimensional P is ϱ-denser than all $P \in \mathcal{P}(L)$.

All ideal crystals can be modeled as periodic packings of spheres in E^3. In Section 4 we choose specific examples with $n = 2$ to show that finite packings equipped with the parametric density is a useful approach to explain crystal growth based on atomic structure only ([6], p.1). We consider crystals of the NaCl-type where K_1 and K_2 are spheres with different radii. For this type M is a lattice again. This does not hold for the ZnS-type, named after the zinc blend. A special case of this type is the diamond which consists of only one type of atoms (carbon). For all examples there are parameters ϱ such that the asymptotic shape of the densest packing coincides with the natural shape, which is the cube for NaCl, the octahedron for the diamond and for the ZnS-type the shape may be a tetrahedron, in particular not centrally symmetric.

2. Bounds for the number of facets

To estimate the number of facets of $W(L,\varrho)$, we first show that only sufficiently small vectors of the polar lattice contribute to the Wulff-shape. In fact we prove this for periodic packings.

Lemma 1. *For $L \in \mathcal{L}^d$ let $f : L^* \to \mathbb{R}$ with $f(nu) = f(u)$, for $n \in \mathbb{N}$, $u \in L^*$ and $0 < a \leq f(u) \leq b$, for all u. Further let $W(L,f,\varrho) = \bigcap_{u \in L^* \, primitive} \{y : uy \leq \varrho\|u\| - f(u)\}$ and $B = \{u \in L^* : f(u) \geq b - a/2\}$ and let there be an $R > 0$ such that $B + RB^d = E^d$. Then for $\varrho > \max_{u \in L^* \, primitive} \left\{\frac{f(u)}{\|u\|}\right\}$ and $S = \max\{2R, 4R^2\varrho/a\}$*

$$W(L,f,\varrho) = \bigcap_{\substack{u \in L^* \, primitive \\ \|u\| \leq S}} \{y : uy \leq \varrho\|u\| - f(u)\}.$$

In particular $W(L,f,\varrho)$ is a polytope.

In the case that even $\{u \in L^* : f(u) = b\} + RB^d$ is a covering, we may choose $S = \max\{2R, 2R^2\varrho/a\}$.

Proof. Let W' be the set on the right side. Then it suffices to prove that $W' \subset W := W(L,f,\varrho)$. Assume $x \in W' \backslash W$. Then there is a $u \in L^*$ with $\|u\| > S$ and $ux > \varrho\|u\| - f(u)$ and without restriction we may assume that $\|u\|$ is minimal. By assumption there is a $v \in B$ with $\|v - \frac{u}{2}\| \leq R$. With $w := u - v$ we have $v + w = u$, $\|w - \frac{u}{2}\| \leq R$ and hence $\|v - w\| \leq 2R$. From $\|v\|, \|w\| \leq \|u/2\| + R < \|u\|$ it follows by the minimal property of u that $vx \leq \varrho\|v\| - f(v)$ and $wx \leq \varrho\|w\| - f(w)$ and so

$$ux \leq \varrho(\|v\| + \|w\|) - b + a/2 - f(w).$$

The parallelogram identity leads to

$$\begin{aligned}(\|v\| + \|w\|)^2 &= 2\|v\|^2 + 2\|w\|^2 - (\|v\| - \|w\|)^2 \\ &\leq 2\|v\|^2 + 2\|w\|^2 = \|v + w\|^2 + \|v - w\|^2 \leq \|u\|^2 + 4R^2\end{aligned}$$

and hence

$$\|v\| + \|w\| \leq \|u\| + \left(\sqrt{\|u\|^2 + 4R^2} - \|u\|\right) = \|u\| + \frac{4R^2}{\|u\| + \sqrt{\|u\|^2 + 4R^2}} \leq \|u\| + \frac{2R^2}{\|u\|}.$$

With $\|u\| > 4R^2\varrho/a$ and $f(u) \leq b$ we get

$$ux \leq \varrho\|u\| + \frac{2\varrho R^2}{\|u\|} - b + a/2 - f(w) \leq \varrho\|u\| + a - f(w) - f(u) \leq \varrho\|u\| - f(u),$$

which contradicts our assumption. The additional remark follows from $f(v) = b$. □

Remark. For the Wulff-shape in the lattice case (4) we have $a = b = 1/2$ and one can choose R equal to the covering radius $\mu(L^*)$. If $N(L,\varrho)$ denotes the number of facets of $W(L,\varrho)$ then by Lemma 1 and $\mu(L^*)\varrho_L = \mu(L^*)/(2\lambda_1(L^*)) \geq 1/4$

$$N(L,\varrho) \leq G(8\mu(L^*)^2\varrho \cdot B^d, L^*), \quad \text{for } \varrho > \varrho_L. \tag{12}$$

We also get lower bounds and we so obtain

Theorem 1. *Let $\varrho \geq 2\varrho_L$. Then*

$$c_1(L)\varrho^{d(d-1)/(d+1)} \leq N(L,\varrho) \leq c_2(L)\varrho^d,$$

where $c_1(L) = \left(\frac{d\kappa_d}{2^d\kappa_{d-1}}\right)^{2d/(d+1)} \left(\frac{\lambda_1(L^*)}{3}\right)^{d(d-1)/(d+1)}$ and $c_2(L) = \kappa_d \det L 12^d \mu(L^*)^{2d}$ depend only on L.

Remarks. (1) Actually the proof shows that for $\varrho > \varrho_L$

$$c_1(L)\left(\frac{4(\varrho - \varrho_L)^2}{\varrho}\right)^{d(d-1)/(d+1)} \leq N(L,\varrho) \leq c_2(L)\varrho^d.$$

(2) Theorem 1 can be interpreted as a stability result on the Wulff-shape depending on ϱ (cf. [9]).

Proof. There are two simple but useful upper bounds for the lattice point enumerator (cf. [8]):

$$\det\Lambda \cdot G(K,\Lambda) \leq V(K + \mu(\Lambda)B^d) \tag{13}$$

and, if Λ is a packing lattice for B^d,

$$\kappa_d \cdot G(K,\Lambda) \leq V(K + B^d). \tag{14}$$

For $K = rB^d$ in (13) and (14) and with $G(rB^d,\Lambda) = G(2r/\lambda_1(\Lambda) \cdot B^d, 2/\lambda_1(\Lambda) \cdot \Lambda)$ in (14) we get

$$G(rB^d,\Lambda) \leq \frac{\kappa_d}{\det\Lambda}(r + \mu(\Lambda))^d \tag{15}$$

and

$$G(rB^d,\Lambda) \leq \left(\frac{2r}{\lambda_1(\Lambda)} + 1\right)^d. \tag{16}$$

Now the upper bound is a consequence of (12), (15) and $1 < 4\mu(L^*)\varrho$, for $\varrho > \varrho_L$.

For the lower bound let $L^*\backslash\{0\} = (u^1, u^2, \dots)$ with $\|u^i\| \le \|u^{i+1}\|$, $i = 1, 2, \dots$. Further let $r_i = \|u^i\|$. From (16) and $r_i \ge \lambda_1(L^*)$ there is a constant $\eta_d = \left(\frac{3}{\lambda_1(L^*)}\right)^d$, such that $G(r_iB^d, L^*) \le \eta_d \cdot r_i^d$. Consequently

$$r_i \ge \eta_d^{-1/d} \cdot G(r_iB^d, L^*)^{1/d} \ge \eta_d^{-1/d} i^{1/d}.$$

Now let $v^1, \dots, v^N \in L^*$ be the lattice vectors corresponding to the $N = N(L, \varrho)$ facets of $W(L, \varrho)$ with $\|v^i\| \le \|v^{i+1}\|$, $i = 1, \dots, N-1$. It follows $\|v^i\| \ge r_i \ge \eta_d^{-1/d} i^{1/d}$, so the i-th facet has distance $\varrho - \frac{1}{2\|v^i\|} \ge \varrho - \frac{\eta_d^{1/d}}{2i^{1/d}}$ to the origin. Hence its $(d-1)$-volume f_i is bounded above by

$$f_i \le \kappa_{d-1} \cdot \left(\varrho^2 - \varrho^2 - \frac{\eta_d^{2/d}}{4i^{2/d}} + \frac{\varrho\eta_d^{1/d}}{i^{1/d}}\right)^{(d-1)/2} \le \kappa_{d-1} \left(\frac{\eta_d}{i}\right)^{(d-1)/(2d)} \varrho^{(d-1)/2}.$$

Consequently, we obtain for the surface area F of $W(L, \varrho)$ the inequality $F \le \kappa_{d-1}\eta_d^{(d-1)/(2d)} \varrho^{(d-1)/2} \sum_{i=1}^N \frac{1}{i^{(d-1)/(2d)}}$.
Since $\sum_{i=1}^N i^{(1-d)/(2d)} \le 1 + \int_1^N x^{(1-d)/(2d)}\,dx = 1 - 2d/(d+1) + 2d/(d+1)N^{(d+1)/(2d)} \le 2N^{(d+1)/(2d)}$ we have

$$F \le 2\kappa_{d-1}\eta_d^{(d-1)/(2d)} \varrho^{(d-1)/2} N^{(d+1)/(2d)}. \tag{17}$$

For the inradius r of $W(L, \varrho)$ holds $r \ge \varrho - 1/(2\lambda_1(L^*)) = \varrho - \varrho_L$ and with $\varrho \ge 2\varrho_L$ it follows

$$F \ge d\kappa_d(\varrho - \varrho_L)^{d-1} \ge \frac{d\kappa_d}{2^{d-1}}\varrho^{d-1}. \tag{18}$$

From (17) and (18) we obtain

$$N \ge \left(\frac{d\kappa_d}{2^d\kappa_{d-1}}\right)^{2d/(d+1)} \eta_d^{-(d-1)/(d+1)} \cdot \varrho^{d(d-1)/(d+1)}$$

and Theorem 1 is proved. □

Example. For $\mathbb{Z}^d$ the constants in Theorem 1 are $c_1 = \left(\frac{3d^2\kappa_d^2}{12^d\kappa_{d-1}^2}\right)^{d/(d+1)} \sim \left(\frac{6\pi d}{12^d}\right)^{d/(d+1)}$ and $c_2 = \kappa_d(3d)^d \sim (18\pi e d)^{d/2}$.

For applications to crystals we can assume that M is rational with respect to L, i.e. there is a lattice Λ such that $M \subset \Lambda$. So we can apply Lemma 1 with $R = \mu(\Lambda^*)$ and we get an upper bound for the number of facets $N(M, \varrho)$ of $W(M, \varrho)$ as in Theorem 1. Since the proof for the lower bound works for all polytopes $P \in \mathcal{P}(L)$ with $W(L, \varrho) \subset P \subset \varrho B^d$, it follows with $0 \le \tau(u) \le 1/2$ (compare Lemma 2 in Section 3)

Corollary 1. *If $M \subset \Lambda$, for a lattice Λ, then there are constants $c_1(M), c_2(M)$ such that for $\varrho \ge 2\varrho_L$*

$$c_1(M)\varrho^{d(d-1)/(d+1)} \le N(M, \varrho) \le c_2(M)\varrho^d.$$

3. General periodic packings

The following explicit representation will later be used to minimize $\Delta_M(P, \varrho)$.

Theorem 2. *Let $P \in \mathcal{P}(L)$ be with outer facet normals $u_i \in L^*$, $i = 1, \dots, k$ and let $f_1, \dots, f_k$ denote the $(d-1)$-dimensional volumes of the corresponding facets $F_1, \dots, F_k$. Then*

$$\Delta_M(P, \varrho) = \delta_M V(P)^{1/d-1} \sum_{i=1}^{k} f_i \left(\varrho - \frac{\tau(u_i)}{\|u_i\|} \right).$$

Proof. Let $\Delta_1, \dots, \Delta_k$ be the $(d-1)$-dimensional determinants of the sublattices of L orthogonal to $u_1, \dots, u_k$. Further let $\{R_\lambda\}$ be a sequence as in Definition 1. In particular, R_λ has the same outer facet normals as P. Further we can assume that 0 is contained in $\operatorname{int}(P)$ and $\operatorname{int}(R_\lambda)$. For $\lambda \geq 1$ and $j = 1, \dots, n$ we define $P_j(\lambda)$ by

$$P_j(\lambda) = \operatorname{conv}(\lambda R_\lambda \cap L_j). \tag{19}$$

To determine $L_j(\lambda R_\lambda) = L(Q_j(\lambda))$, where $Q_j(\lambda) = P_j(\lambda) - t_j$, we consider the contribution of each lattice point $u \in L$ to $L(Q_j(\lambda)) - V(Q_j(\lambda))/\det L$. If $D(u)$ denotes the Dirichlet-Voronoi-cell (DV-cell) corresponding to $u \in L$, then we distinguish five cases:

(1) $D(u) \subset Q_j(\lambda)$,
(2) $D(u) \subset E^d \backslash Q_j(\lambda)$,
(3) u is contained in a facet of $Q_j(\lambda)$,
(4) $u \in \operatorname{int} Q_j(\lambda)$ and $D(u) \not\subset Q_j(\lambda)$,
(5) $u \notin Q_j(\lambda)$ and $D(u) \cap Q_j(\lambda) \neq \emptyset$.

We consider the lattice points in four steps.

(a) The contribution of the $u \in L$ of types (1) and (2) is 0.

From $R_\lambda \to P$ it is clear that the facets $G_{\lambda,i}$ of R_λ orthogonal to u_i have $(d-1)$-volume $f_i \lambda^{d-1} + o(\lambda^{d-1})$. The convex hull of the points of L_j in the parallel L_j-hyperplane close to $G_{\lambda,i}$ defines facets $F_{\lambda,i}$ of $Q_j(\lambda)$ orthogonal to u_i with $(d-1)$-volume $V_{d-1}(G_{\lambda,i}) + O(\lambda^{d-2}) = f_i \lambda^{d-1} + o(\lambda^{d-1})$. Let $H_{\lambda,i} = \operatorname{aff}(F_{\lambda,i})$, $Q_j(\lambda)$ contained in the halfspace $H^+_{\lambda,i}$ and $\Lambda_{\lambda,i} = L \cap H_{\lambda,i}$. For $u \in \Lambda_{\lambda,i}$ let $D_{d-1}(u)$ be the corresponding $(d-1)$-dimensional DV-cell. Considering the lattice points in the inner parallel body of $F_{\lambda,i}$ in $H_{\lambda,i}$ with a distance, which depends only on L and P, it follows that there are $\frac{f_i}{\Delta_i} \lambda^{d-1} + o(\lambda^{d-1})$ DV-cells $D_{d-1}(u)$ of $\Lambda_{\lambda,i}$ with $((D_{d-1}(u) + 2D(0)) \cap H^+_{\lambda,i}) \subset Q_j(\lambda)$.

(b) All these lattice points $u \in \Lambda_{\lambda,i}$ (of type (3)) have a contribution of $1/2$.

(c) Next we show that the (negative) contribution of lattice points of type (5) whose DV-cells intersect one of these $(d-1)$-cells is canceled out by those of type (4). It suffices to consider one such $u \in \Lambda_{\lambda,i}$. Let $v \in L$ of type (5) be with $D(v) \cap D_{d-1}(u) \neq \emptyset$. By the choice of u we have $(D(2u - v) \cap H_i^+) \subset Q_j(\lambda)$ and the sum of the contribution of v and its mirror point $2u - v$ is 0.

(d) For the surface area of $Q_j(\lambda)$ it holds $F(Q_j(\lambda)) = \sum_{i=1}^k f_i \lambda^{d-1} + o(\lambda^{d-1})$ and hence the remaining parts of the boundary of $Q_j(\lambda)$ have a total $(d-1)$-volume of $o(\lambda^{d-1})$ and so the total contribution of DV-cells intersecting those parts is $o(\lambda^{d-1})$.

Combining (a), (b), (c) and (d) we obtain (analogously to Ehrhart's formula for the lattice point enumerator (cf. e.g. [10], [8]))

$$L_j(\lambda R_\lambda) = L(Q_j(\lambda)) = \frac{V(P_j(\lambda))}{\det L} + 1/2\lambda^{d-1}\sum_{i=1}^{k}\frac{f_i}{\Delta_i} + o(\lambda^{d-1}), \tag{20}$$

for $j = 1, \ldots, n$. Now we compare λR_λ with $P_j(\lambda)$. We dissect λR_λ into the cones $C_i = \mathrm{pos}\,(F_i)$. Let $c_i(\lambda) = \max\left\{\frac{u_i}{\|u_i\|}x : x \in \lambda R_\lambda \cap M\right\}$, $c_{ij}(\lambda) = \max\left\{\frac{u_i}{\|u_i\|}x : x \in \lambda R_\lambda \cap L_j\right\}$ and $\alpha_{ij}(\lambda) = c_i(\lambda) - c_{ij}(\lambda) \geq 0$. The contribution to $V(\lambda R_\lambda) - V(P_j(\lambda))$ is $\alpha_{ij}(\lambda) f_i \lambda^{d-1} + o(\lambda^{d-1})$. Summing up over all facets yields

$$V(P_j(\lambda)) = V(\lambda R_\lambda) - \lambda^{d-1}\sum_{i=1}^{k}\alpha_{ij}(\lambda) f_i + o(\lambda^{d-1}).$$

Together with (20) and $\Psi(R_\lambda, \lambda) = \sum_{j=1}^{n} v_j L_j(\lambda R_\lambda)$ it follows

$$\begin{aligned}\Psi(R_\lambda, \lambda) &= V(\lambda R_\lambda)\frac{v}{\det L} + \lambda^{d-1}\sum_{i=1}^{k}\sum_{j=1}^{n} f_i v_j\left(\frac{1}{2\Delta_i} - \frac{\alpha_{ij}(\lambda)}{\det L}\right) + o(\lambda^{d-1}) \\ &= \delta_M V(\lambda R_\lambda) + \delta_M \lambda^{d-1}\sum_{i=1}^{k} f_i \frac{\det L}{\Delta_i}\left(\frac{1}{2} - \sum_{j=1}^{n}\frac{\Delta_i}{\det L}\alpha_{ij}(\lambda)\frac{v_j}{v}\right) + o(\lambda^{d-1}).\end{aligned}$$

Without restriction we can assume that there is an index $m = m(\lambda)$ with $\alpha_{im}(\lambda) = 0$ and hence $\frac{\Delta_i}{\det L}\alpha_{ij}(\lambda) = \epsilon_m(u_i) - \epsilon_j(u_i)$. From the definition of $\tau_m(u_i)$ and $\tau(u_i)$ together with $\Delta_i = \det L\|u_i\|$ we obtain

$$\Psi(R_\lambda, \lambda) \leq \delta_M V(\lambda R_\lambda) + \delta_M \lambda^{d-1}\sum_{i=1}^{k} f_i \frac{\tau(u_i)}{\|u_i\|} + o(\lambda^{d-1}).$$

Now by Definition 1 and $R_\lambda \to P$

$$\begin{aligned}\Delta_M(P, \varrho) &\geq \delta_M \lim_{\lambda\to\infty} V(\lambda R_\lambda)^{1/d} \\ &\quad \left(\frac{V(\lambda R_\lambda) + \varrho F(\lambda R_\lambda) - V(\lambda R_\lambda) - \lambda^{d-1}\sum_{i=1}^{k} f_i \frac{\tau(u_i)}{\|u_i\|} + o(\lambda^{d-1})}{V(\lambda R_\lambda) + o(\lambda^d)}\right) \\ &= \delta_M \lim_{\lambda\to\infty} V(R_\lambda)^{1/d-1}\left(\varrho F(R_\lambda) - \sum_{i=1}^{k} f_i \frac{\tau(u_i)}{\|u_i\|}\right) \\ &= \delta_M V(P)^{1/d-1}\sum_{i=1}^{k} f_i\left(\varrho - \frac{\tau(u_i)}{\|u_i\|}\right)\end{aligned}$$

On the other hand we can choose R_λ such that $\tau(u_i)$ is attained for $i = 1, \ldots, k$ and equality follows. □

Remark. If in the definition of the parametric density (5) B^d is replaced by a convex body C, then a density deviation $\Delta_M(P, C, \varrho)$ with respect to C can be defined. The same proof as in Theorem 2 yields

$$\Delta_M(P,C,\varrho) = \delta_M V(P)^{1/d-1} \sum_{i=1}^{k} \frac{f_i}{\|u_i\|} \left(\varrho h_C(u_i) - \tau(u_i)\right), \qquad \square$$

where h_C is the support function of C.

Proposition 1. *Let $P, Q \in \mathcal{P}(L)$ and $\varrho > \varrho_M$ with $\Delta_M(P,\varrho) < \Delta_M(Q,\varrho)$. Then P is asymptotically ϱ-denser than Q.*

Proof. The proof of Theorem 2 shows that there is a sequence $R_\lambda \in \mathcal{P}(P)$ with $R_\lambda \to P$ and a constant c with

$$V(\lambda R_\lambda) \le V((\lambda+1)R_{\lambda+1}) \le V(\lambda R_\lambda) + c\lambda^{d-1}$$

and

$$\delta_\varrho(M, \lambda R_\lambda) = \delta_M - \frac{\Delta_M(P,\varrho)}{V(\lambda R_\lambda)^{1/d}} + o(1/\lambda).$$

On the other hand, by the definition of Δ_M for any sequence $S_\nu \in \mathcal{P}(Q)$ with $S_\nu \to Q$ holds

$$\delta_\varrho(M, \nu S_\nu) \le \delta_M - \frac{\Delta_M(Q,\varrho)}{V(\nu S_\nu)^{1/d}} + o(1/\nu).$$

Now we define $\lambda(\nu)$ by

$$M(\lambda(\nu)R_{\lambda(\nu)}) \le M(\nu S_\nu) \le M((\lambda(\nu)+1)R_{\lambda(\nu)+1}).$$

From $M(\nu S_\nu) = nV(\nu S_\nu)/\det L + o(\nu^d)$ and $M(\lambda(\nu)R_{\lambda(\nu)}) = nV(\lambda(\nu)R_{\lambda(\nu)})/\det L + o(\lambda(\nu)^d)$ it follows $V(\lambda(\nu)R_{\lambda(\nu)}) = V(\nu S_\nu) + o(\nu^d)$ and $\lambda(\nu) = \Omega(\nu)$. Hence

$$\begin{aligned}
\delta_\varrho(M, \lambda(\nu)R_{\lambda(\nu)}) - \delta_\varrho(M, \nu S_\nu) &\ge \frac{\Delta_M(Q,\varrho) - \Delta_M(P,\varrho)}{V(\nu S_\nu)^{1/d}} + o(1/\nu) \\
&= 1/\nu \cdot \left(\frac{\Delta_M(Q,\varrho) - \Delta_M(P,\varrho)}{V(Q)^{1/d}}\right) + o(1/\nu)
\end{aligned}$$

and the assertion is a consequence of $\delta_\varrho(M, \lambda R_\lambda) - \delta_\varrho(M, (\lambda+1)R_{\lambda+1}) = o(1/\lambda)$. $\square$

To prove that W_M is a polytope we first have to check that the assumptions of Lemma 1 are satisfied.

Lemma 2. *For $L \in \mathcal{L}^d$ and $u \in L^*$*

$$\frac{1}{2n} \le \tau(u) \le \frac{1}{2}.$$

Proof. The right inequality is clear and equality holds if M is a lattice. For the lower bound let $\lambda_j = v_j/v$, $j = 1, \ldots, n$. Then $\sum_{j=1}^n \lambda_j = 1$ and hence $\sum_{j=1}^n \lambda_j^2 \ge 1/n$. With $\{\epsilon_m - \epsilon_j\} + \{\epsilon_j - \epsilon_m\} \le 1$ (we omit the (u)) we get

$$\begin{aligned}
\sum_{m=1}^n \lambda_m \sum_{j=1}^n \lambda\{\epsilon_m - \epsilon_j\} &= \sum_{m \ne j} \lambda_j \lambda_m \{\epsilon_m - \epsilon_j\} \\
&\le \sum_{j<m} \lambda_j \lambda_m = \frac{1}{2}\left(1 - \sum_{j=1}^n \lambda_j^2\right) \le \frac{1}{2} - \frac{1}{2n}.
\end{aligned}$$

Hence there is an $m \in \{1, \dots n\}$ with $\sum_{j=1}^{n} \lambda_j \{\epsilon_m - \epsilon_j\} \leq 1/2 - 1/(2n)$ and so $\tau(u) \geq 1/(2n)$. Equality holds for $\lambda_j = 1/n$ and $\epsilon_j = (j-1)/n,\ j = 1, \dots, n$. □

From Corollary 1 we know that W_M is a polytope, if M is rational with respect to L. This is sufficient for application to crystals but for completeness we also consider the irrational case.

Theorem 3. *If $\varrho > \varrho_M$, then W_M is a polytope with nonempty interior, i.e. $W_M \in \mathcal{P}(L)$.*

Proof. From the definitions of W_M and ϱ_m we have $0 \in \operatorname{int}(W_M)$. In Lemma 1 we have $f(u) = \tau(u)$, $a = 1/(2n), b = 1/2$ and $B = \{u \in L^* : \tau(u) \geq 1/2 - 1/(4n)\}$. We have to prove the existence of an $R > 0$ with $RB^d + B = E^d$. For this let $L = A\mathbb{Z}^d, m \in L^*$, $\alpha_j = t_j m,\ j = 1, \dots, n$. We define homogenous linear forms $L_1, \dots, L_n$ by $L_j(x) = t_j A^* x$, where $A^* = (A^{-1})^t$. If $\sum_{j=1}^{n} w_j L_j(x)$ has integer coefficients, then $\sum_{j=1}^{n} w_j t_j \in L$ and $\sum_{j=1}^{n} w_j \alpha_j = \left(\sum_{j=1}^{n} w_j t_j\right) m \in \mathbb{Z}$. It follows by Kronecker's approximation theorem (see e.g. [3], p.58), that there is a $v \in \mathbb{Z}^d$ and an $X > 0$ such that

$$\|L_j(v) - \alpha_j\|_{\mathbb{Z}} < \frac{1}{8n}, \quad j = 1, \dots, n$$

and

$$|v_i| \leq X, \quad i = 1, \dots, d,$$

where $\|\cdot\|_{\mathbb{Z}}$ denotes the distance to $\mathbb{Z}$. With $u = A^* v - m \in L^*$ we obtain $\|ut_j\|_{\mathbb{Z}} < 1/(8n)$ and $\|u + m\| \leq \|A^*\|\sqrt{d}X$. Hence $\tau(u) > 1/2 - 1/(4n)$ and the assertion is a consequence of $R = \|A^*\|\sqrt{d}X + \mu(L^*)$. □

We will prove that W_M minimizes $\Delta_M(P, \varrho)$ for all $P \in \mathcal{P}(L)$ (note that $\Delta_M(P, \varrho)$ is invariant for dilatations of P). In fact we prove a more general result.

Lemma 3. *Let $u_1, u_2, \dots \in E^d$ and $\alpha_1, \alpha_2, \dots \in \mathbb{R}$, such that*

$$W = \bigcap_{i=1}^{\infty} \{y : u_i/\|u_i\| y \leq \alpha_i\}$$

is a polytope with $\operatorname{int}(W) \neq \emptyset$. Then holds for all polytopes Q with facet normals from $\{u_1, u_2, \dots\}$

$$V(Q)^{1/d-1} \sum \alpha_i f_i(Q) \geq dV(W)^{1/d},$$

with equality if and only if Q is homothetic to W.

Remark. Actually Lemma 3 is Wulff's theorem (compare [5], [7], [11], [17]).

Proof. Let h be the support function of W. Then it follows from the definition of W that

$$h(u_i/\|u_i\|) \leq \alpha_i, \quad \text{for all } i.$$

We can estimate $\sum \alpha_i f_i(Q)$ with the help of a mixed volume (see [13]) and Minkowski's inequality (see [13]) yields that

$$\begin{aligned} V(Q)^{1/d-1} \sum \alpha_i f_i(Q) &\geq V(Q)^{1/d-1} \sum f_i(Q) h(u_i/\|u_i\|) = V(Q)^{1/d-1} dV_{d-1}(W, Q) \\ &\geq V(Q)^{1/d-1} dV(W)^{1/d} V(Q)^{(d-1)/d} = dV(W)^{1/d} \end{aligned}$$

with equality only if Q and W are homothetic. On the other hand, for $Q = W$ equality holds. □

An immediate consequence is

Corollary 2. *If $V(Q) = V(W_M)$, for any $Q \in \mathcal{P}(L)$, then $\Delta_M(Q, \varrho) \geq \Delta_M(W_M, \varrho)$, with equality only if Q is a translate of W_M.*

Remark. With C instead of B^d in (5) we obtain the Wulff-shape

$$W(M, \varrho, C) = \{x : ux \leq \varrho h_C(u) - \tau(u),\ u \in L^* \text{ primitive}\}.$$

4. Application to crystal types

We consider some examples of crystal-types, which can be described as two-lattice packings. For $n = 2$ we assume $v_1 \geq v_2$. Let $\sigma = \frac{v_1}{v_1+v_2} \in [1/2, 1]$. Actually, σ is not well-defined in reality. But we can consider it as an average value. For shortness let $t = t_2$ and $\epsilon(u) = \epsilon_2(u)$, for $u \in L^*$. Consequently $\tau(u) = 1/2 - \min\{\epsilon(u)\sigma, (1 - \epsilon(u))(1 - \sigma)\}$.

First we consider crystals of the NaCl-type, for which $M = \mathbb{Z}^3$ is a lattice again. The bigger atomic ions (Na) are placed in the even points (i.e. with even sum of coordinates) and the smaller ones (Cl) in the odd points. It is well-known that the odd and the even points in $\mathbb{Z}^3$ are copies of the face centered cubic (fcc) lattice with basis $\{(1,1,0)^t, (1,0,1)^t, (0,1,1)^t\}$. The translation vector is $t = (1,0,0)^t$. We have $\det L = 2$, and L^* has a basis $\{1/2(1,1,-1)^t, 1/2(1,-1,1)^t, 1/2(-1,1,1)^t\}$. Further $\epsilon(u) \in \{0, 1/2\}$ for $u \in L^*$, and hence $\epsilon(-u) = \epsilon(u)$ and $\varrho_M = \max\{1/2, \sigma/\sqrt{3}\}$.

We determine $W(M, \varrho)$ depending on ϱ and σ. For large ϱ the shape will tend to a ball and exact computation is hard. But for small ϱ we obtain the natural shapes.

Theorem 4. *Let $\varrho > \varrho_M = \max\{1/2, \sigma/\sqrt{3}\}$. Then $W(M, \varrho)$ is*

(a) *a cube of edgelength $(2\varrho - 1)$, if $1/2 \leq \varrho \leq 1/12(3 + \sqrt{3})(3 - 2\sigma)$.*

(b) *an octahedron of height $2(\varrho\sqrt{3} - \sigma)$, if $\sigma/\sqrt{3} \leq \varrho \leq 1/2(1 + \sqrt{3})(\sigma - 1/2)$.*

(c) *a truncated octahedron, if $\max\{(2 + \sqrt{3})(1 - \sigma), 1/2(1 + \sqrt{3})(\sigma - 1/2)\} \leq \varrho \leq (\sqrt{2} + \sqrt{3})(\sigma - 1/2)$.*

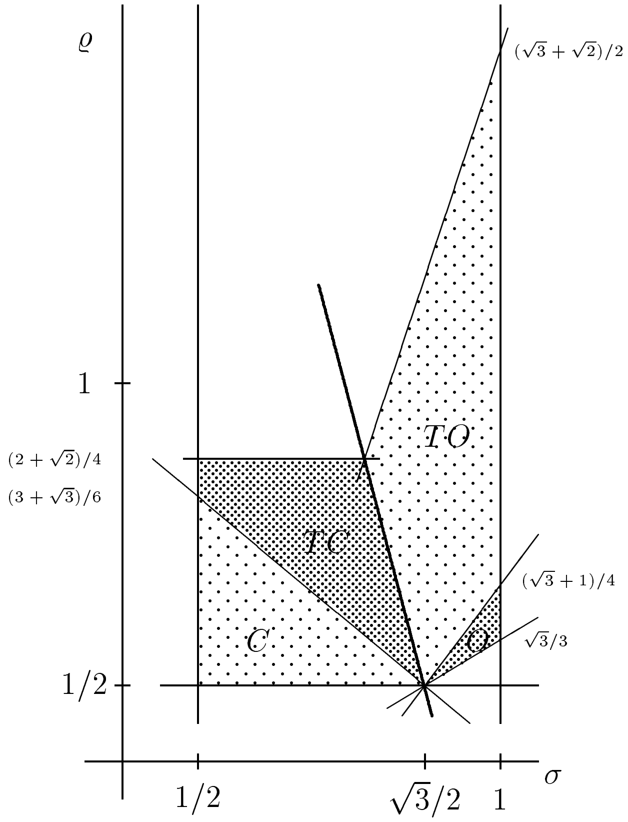
(d) *a truncated cube, if $1/12(3 + \sqrt{3})(3 - 2\sigma) \leq \varrho \leq \min\{(2 + \sqrt{3})(1 - \sigma), 1/4(2 + \sqrt{2})\}$.*

Remark. We illustrate the theorem in the following diagram (see below).

For example $r(Na^+) = 116\text{pm}$, $r(Cl^-) = 167\text{pm}$ (see e.g. [4]) and $\sigma = 0.748\ldots$. So the Wulff-shape for NaCl is a cube for $\varrho \in (0.5, 0.592\ldots)$.

Proof. (a) We have $\pm(1,0,0)^t, \pm(0,1,0)^t, \pm(0,0,1)^t \in L^*$ with $\epsilon = 0$. The intersection of the corresponding halfspaces yields the cube C. By symmetry it suffices to prove that $x_0 = (\varrho - 1/2)(1,1,1)^t$ satisfies the conditions for all $u \in L^*$. Therefore by assumption on ϱ

$$\begin{aligned} ux_0 &\leq \|u\|\|x_0\| = (\varrho - 1/2)\sqrt{3}\|u\| = \varrho\|u\| + (\varrho(\sqrt{3} - 1) - \sqrt{3}/2)\|u\| \\ &\leq \varrho\|u\| + (1/12(3 + \sqrt{3})(3 - 2\sigma)(\sqrt{3} - 1) - \sqrt{3}/2)\|u\| \\ &\leq \varrho\|u\| - \sigma/\sqrt{3}\|u\|. \end{aligned}$$



For those u with $\epsilon(u) = 1/2$ the desired inequality is $ux_0 \leq \varrho\|u\| - \sigma/2$ and is satisfied since $\|u\| \geq \sqrt{3}/2$, for $u \in L^*$. For $\epsilon(u) = 0$, $\|u\| \geq \sqrt{3} \geq \sqrt{3}/(2\sigma)$ is sufficient. So we still have to check the twelve $u \in L^*$ with $\|u\| = \sqrt{2}$ and obviously it suffices to consider $u = (1,1,0)^t$. By assumption

$$ux_0 = 2\varrho - 1 \leq \varrho\sqrt{2} + (2-\sqrt{2})1/12(3+\sqrt{3})(3 - 2\cdot 1/2) - 1 < \varrho\sqrt{2} - 1/2.$$

(b) The 8 vectors in L^* with length $\sqrt{3}/2$ determine the octahedron O. Again it suffices to prove that $x_0 = (\varrho\sqrt{3} - \sigma)(1,0,0)^t$ satisfies the conditions for all $u \in L^*$. All further $u \in L^*$ have length ≥ 1 and by our assumption on ϱ

$$\begin{aligned} ux_0 &\leq (\varrho\sqrt{3} - \sigma)\|u\| \leq \varrho\|u\| + \Big(1/2(1+\sqrt{3})(\sigma - 1/2)(\sqrt{3}-1) - \sigma\Big)\|u\| \\ &= \varrho\|u\| - \|u\|/2 \leq \varrho\|u\| - 1/2. \end{aligned}$$

(c) Let $a = \varrho\sqrt{3} - \sigma$ and $b = \varrho - 1/2$. Then $a, b \geq 0$. The six vectors in L^* with length 1 and $\epsilon = 0$ determine a cube C with height $2b$ and the eight vectors in L^* with length $\sqrt{3}/2$ and $\epsilon = 1/2$ determine an octahedron O with height $2a$. By the assumptions on ϱ and σ we have $b \leq a \leq 2b$ and the intersection of O and C is a truncated octahedron with 24 vertices of the type $v = (0, a-b, b)^t$. The scalar product of v with one of the six $u \in L^*$ with length $\sqrt{2}$ is at most a and the third assumption on ϱ is equivalent to $a \leq \varrho\sqrt{2} - 1/2$, which is the desired inequality for u. All further primitive $u \in L^*$ have length $\geq \sqrt{11}/2$ and by our assumption $\sigma \geq 1/2 + \varrho(\sqrt{3} - \sqrt{2})$ and hence $a - b \leq \varrho(\sqrt{2} - 1)$. Together with $a - b \leq b = \varrho - 1/2$ one obtains by elementary calculation that $\|v\| \leq \varrho - 1/\sqrt{11}$ and hence $uv \leq \varrho\|u\| - \|u\|/\sqrt{11} \leq \varrho\|u\| - 1/2$.

(d) Let a, b be as in (c). By the assumptions on ϱ and σ we have $2b \leq a \leq 3b$ and here the intersection of O and C is a truncated cube with 24 vertices of the type $v = (b, b, a - 2b)^t$. The scalar product of v with one of the six $u \in L^*$ with length $\sqrt{2}$ is at most $2b$ and the third assumption on ϱ is equivalent to $2b \leq \varrho\sqrt{2} - 1/2$, which is the desired inequality for u. The scalar product of v with one of the 24 $u \in L^*$ with length $\sqrt{11}/2$, e.g. $1/2(3, 1, 1)^t$ is at most $5b/2 = 5/2\varrho - 5/4 \leq \varrho\sqrt{11}/2 - 1/2$. All further primitive $u \in L^*$ have length $\geq \sqrt{19}/2$. Further, $\|v\| \leq b\sqrt{3}$ and hence

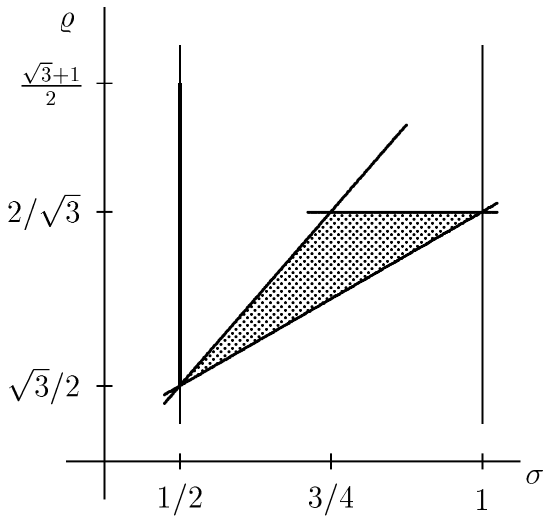
$$uv \leq \varrho\|u\| + \left(\varrho(\sqrt{3} - 1) - \sqrt{3}/2 - \varrho\right)\|u\| \leq \varrho\|u\| - 1/2. \qquad \square$$

Now we turn to crystals of the ZnS-type, named after the zinc blend. One special case of this type is the diamond, which consists of only one type of atoms (carbon) and consequently $\sigma = 1/2$. Again L is a face centered cubic lattice (with basis $\{(2, 2, 0)^t, (2, 0, 2)^t, (0, 2, 2)^t\}$) and the translation vector is $t = (1, -1, 1)^t$. We have $\det L = 16$, and L^* has a basis $\{1/4(1, 1, -1)^t, 1/4(1, -1, 1)^t, 1/4(-1, 1, 1)^t\}$. Further $\epsilon(u) \in \{0, 1/4, 1/2, 3/4\}$, for $u \in L^*$ and $\varrho_M = (\sigma + 1)/\sqrt{3}$.

Theorem 5. (a) *If $\sqrt{3}/2 \leq \varrho \leq 1/2(\sqrt{3} + 1)$ and $\sigma = 1/2$, then $W(M, \varrho)$ is an octahedron O with height $2\varrho\sqrt{3} - 3$.*

(b) *If $\frac{\sigma+1}{\sqrt{3}} < \varrho < \min\{\frac{4\sigma+1}{2\sqrt{3}}, \frac{2}{\sqrt{3}}\}$, then $W(M, \varrho)$ is a tetrahedron with outer normal vectors $(-1, -1, -1)^t, (1, 1, -1)^t, (1, -1, 1)^t, (-1, 1, 1)^t$.*

Remark. We illustrate the theorem in the following diagram.



For example $r(Zn^{2+}) = 88$pm, $r(S^{2-}) = 170$pm (see e.g. [4]) and $\sigma = 0.878\ldots$. So the Wulff-shape for ZnS is a tetrahedron for $\varrho \in (1.084\ldots, 1.154\ldots)$.

Remark. Note that in the case of the diamond (a) the planes determining the octahedron are not those with the maximal density (these are the planes with normals $(1,1,0)^t$ etc., which contain both types of atoms). The result is based on the fact that in directions $(1,1,1)$ etc. there are pairs of planes which are very close together.

Proof. (a) For $\sigma = 1/2$ it follows $\tau(u) = 1/2\max\{1-\epsilon(u), \epsilon(u)\} = \tau(-u)$. The eight vectors in L^* with length $\sqrt{3}/4$ and $\tau = 3/8$ determine the octahedron O. It suffices to prove that $x_0 = (\varrho\sqrt{3} - 3/2)(1,0,0)^t$ satisfies the conditions for all further $u \in L^*$. By our assumption on ϱ

$$\begin{aligned} ux_0 &\leq \|u\|(\varrho\sqrt{3} - 3/2) \leq \varrho\|u\| + \left(1/2(1+\sqrt{3})(\sqrt{3}-1) - 3/2\right)\|u\| \\ &= \varrho\|u\| - \|u\|/2. \end{aligned}$$

If $\tau(u) = 1/4$, then $\|u\| \geq 1/2$ and hence $ux_0 \leq \varrho\|u\| - 1/4$.
If $\tau(u) = 3/8$, then (up to the vectors which determine O) $\|u\| \geq \sqrt{11}/4$ and so $ux_0 \leq \varrho\|u\| - \sqrt{11}/8 < \varrho\|u\| - 3/8$.
If $\tau(u) = 1/2$, then for $\|u\| \geq 1$ holds $ux_0 \leq \varrho\|u\| - 1/2$ and we still have to check the u with length $\sqrt{2}/2$. It suffices to consider $u_0 = (1/2, 1/2, 0)^t$:

$$u_0x_0 = \varrho\sqrt{3}/2 - 3/4 \leq \varrho\sqrt{2}/2 + 1/2(\sqrt{3}+1)1/2(\sqrt{3}-\sqrt{2}) - 3/4 < \varrho\sqrt{2}/2 - 1/2.$$

(b) There are four vectors $u = 1/4(-1,-1,-1)^t, 1/4(-1,1,1)^t, 1/4(1,-1,1)^t, 1/4(1,1,-1)^t$ with $\epsilon(u) = 3/4$ and $\tau(u) = (1+\sigma)/4$. The corresponding halfspaces are defined by $(4u)x \leq q := \varrho\sqrt{3} - (\sigma+1)$ and the intersection is a tetrahedron T with vertices $(q,q,q)^t, (-q,q,-q)^t, (q,-q,-q)^t, (-q,-q,q)^t$. We still have to prove that these vertices satisfy the conditions for all the other $u \in L^*$.

For the negatives to the four vectors defining T holds $\epsilon(u) = 1/4$ and $\tau(u) = 1/4\max\{2-\sigma, 3\sigma-1\}$ and the corresponding halfspaces are defined by $(4u)x \leq p := \varrho\sqrt{3} - \max\{3\sigma-1, 2-\sigma\}$. With $p - 3q = -2\varrho\sqrt{3} + \min\{4, 1+4\sigma\}$ it follows by the assumptions on ϱ that $3q \leq p$ and so the vertices of T satisfy these conditions.

If v is a vertex of T and $u \in L^*$ then $uv \leq q\sqrt{3}\|u\| = \varrho\|u\| + (2\varrho - \sqrt{3}(\sigma+1))\|u\|$. The assumptions on ϱ determine a triangle with vertices $(\sigma, \varrho) = (1/2, \sqrt{3}/2), (1, 2/\sqrt{3}), (3/4, \sqrt{3})$. The linear function $(2\varrho - \sqrt{3}(\sigma+1))$ attains its maximum in a vertex and hence $uv \leq \varrho\|u\| - 5\sqrt{3}/12\|u\|$.

For $\|u\| \geq 6/(5\sqrt{3}) = 0.69\ldots$ we have $uv \leq \varrho\|u\| - 1/2$. So we still have to check the six vectors u with length $1/2$ and $\epsilon(u) = 1/2$ and $\tau(u) = \sigma/2$, e.g. $(1/2,0,0)^t$. From $\varrho \leq 2/\sqrt{3}$ we obtain $uv \leq q/2 \leq \varrho\|u\| - \sigma/2$. □

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