Crystalline and Isoperimetric Square Configurations

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We present crystallization results for planar atomic interactions governed by two- and three-body terms with the resulting periodicity being that of the square lattice. The emergence of a (square) Wulff shape for ground states is established by showing the optimality of ground-state configurations in terms of a discrete isoperimetric inequality. Furthermore, an $n^{3/4}$ law for the deviation from the asymptotic Wulff shape is established with an explicit constant for the leading term.

1 Introduction

In this paper the fundamental crystallization problem of analytically explaining why particles at low temperature arrange in periodic lattices is considered. We work in the classical framework and refer to the two-dimensional problem in the square lattice. At low temperatures particle interactions are expected to be essentially determined by particle positions. More precisely, if every particle configuration is identified with the set of its particle positions $x_1, \ldots, x_n$ in $\mathbb{R}^2$, the crystallization problem consists in verifying the periodicity of ground-state configurations of a suitable energy $E : \mathbb{R}^{2n} \rightarrow \mathbb{R} \cup \{+\infty\}$. The energy $E$ is given by the sum of a two-body and a three-body interaction contribution, $E_2$ and $E_3$, respectively.

The literature on two-dimensional crystallization includes [2] as a first result for a two-body sticky interaction energy, inducing triangular lattice periodicity. The result was then extended in [5] to the case of short-ranged soft interactions. The first result accounting for long-range interactions has been instead achieved in [7] where an $E_2$ term of Lennard-Jones type has been considered. Analogous results are established in [1] for the hexagonal lattice by adding a three-body interaction term that favors triples of bonded particles forming bond angles $2\pi/3$ and $4\pi/3$. Furthermore, the emergence of a macroscopic Wulff shape for the triangular lattice with short-ranged two-body interactions has been recently investigated in [6, 8].

In this paper we summarize the results contained in [3]. We consider a short-range $E_2$ term and a $E_3$ term that favors bond angles of $\pi/2$, $\pi$, and $3\pi/2$, with a resulting square ordering (see detailed hypotheses on $E$ in Section 2). Let $\beta(n) := [2n - 2\sqrt{n}]$ where $[\cdot]$ is the right-continuous integer-part $[x] := \max\{z \in \mathbb{Z} : z \leq x\}$. Under the assumptions of Section 2 on the interaction energy, we prove the following main crystallization result.

**Theorem 1.1** Ground states of $E$ are connected subsets of the unit square lattice and their energy is $-\beta(n)$.

As the energy $E$ favors particle clustering and ‘boundary’ particles have in general less bonds, ground states can be intuitively expected to have minimal ‘perimeter’, or maximal ‘area’. This intuition is verified in Section 4 by introducing a suitable notion of perimeter and area of configurations, and by showing that ground states are characterized as those configurations which realize equality in a discrete isoperimetric inequality (see Theorem 4.1). Furthermore, the exact quantification of ground-state perimeter achieved in Theorem 4.1 allows us (still under the assumptions of Section 2) to show the emergence of an asymptotic (square) Wulff shape for ground states and to investigate the ground-state deviation from it. In the following, let us denote by $\mu_{(y_1, \ldots, y_n)}$ the empirical measure $\frac{1}{n} \sum_1 \delta_{y_n/\sqrt{n}}$ of the rescaled configuration $\{y_1/\sqrt{n}, \ldots, y_n/\sqrt{n}\}$.

**Theorem 1.2** Ground states approach the square of side $[\sqrt{n}]$ as the particle number $n$ grows. More precisely, if $\{C_n\}$ is a sequence of ground states and $\mu$ is the Lebesgue measure restricted to the unit square $[0, 1]^2$, then $\{\mu_{C_n}\}$ weak-converges to $\mu$ where each $C_n$ is a suitable rotation and translation of $C_n$. Furthermore, every ground state (up to a rotation and a translation) differs from $S_n := \{(i, j) : i, j = 0, \ldots, \sqrt{n}\}$ by at most $3n^{3/4} + O(n^{1/2})$ particles.

We observe that Theorem 1.2 that is established in Section 4 provides an explicit constant for the leading-order term $n^{3/4}$ of the deviation of ground states from the Wulff shape (see also [6]). Moreover, it easily follows from this result (see in particular (14)) that

$$\|\mu_{C_n} - \mu_{S_n}\| \leq \frac{\#(C_0 \triangle S_n)}{n} \leq 3n^{-1/4} + O(n^{-1/2})$$

where $\|\cdot\|$ stands for the total variation norm. We also have that $\|\mu_{C_n} - \mu\|_F \leq 3n^{-1/4} + O(n^{-1/2})$ where $\|\cdot\|_F$ denotes the flat norm. These results nicely reflect the inherent multiscale nature of the crystallization phenomenon.

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2 Notation and Assumptions on the Interaction Energy

We denote a configuration of $n$ particles by $C_n := \{x_1, \ldots, x_n\} \in \mathbb{R}^{2n}$, the distance between two of its particles, $x_i$ and $x_j$, by $\ell_{ij}$, and the counterclockwise-oriented angle between the two segments $x_i - x_j$ and $x_k - x_j$ by $\theta_{ijk}$. The energy of a configuration $C_n$ is defined by

$$E(C_n) := E_2(C_n) + E_3(C_n) = \frac{1}{2} \sum_{i \neq j} v_2(\ell_{ij}) + \frac{1}{2} \sum_{(i, j, k) \in \mathcal{A}} v_3(\theta_{ijk})$$

(1)

where the functions $v_2 : [0, \infty) \to [-1, \infty]$ and $v_3 : [0, 2\pi) \to [0, \infty)$ are the two-body and the three-body interaction potentials. We choose a strongly-repulsive, short-ranged two-body potential satisfying the following assumptions

$$v_2(\ell) = +\infty \text{ if } \ell < 1,
\hspace{1cm} v_2(\ell) = -1 \text{ if } \ell = 1,
\hspace{1cm} v_2(\ell) = v(\ell) \text{ if } 1 < \ell < \ell^*,
\hspace{1cm} v_2(\ell) = 0 \text{ if } \ell \geq \ell^*,$$

(2)

where $v$ is any function taking values in $(-1, 0)$ and $\ell^* \in (1, \sqrt{2})$ is given. On the other hand, let $\sigma \in (0, \pi/8)$ and define

$$I_1 := [\pi/2 - \sigma, \pi/2 + \sigma],
I_2 := [\pi - \sigma, \pi + \sigma],
I_3 := \left[\frac{3\pi}{2} - \sigma, \frac{3\pi}{2} + \sigma\right],
I := I_1 \cup I_2 \cup I_3.$$

(3)

Let $\theta_{\min} := 2\arcsin(1/(2\sqrt{2})) \approx 0.23\pi$. The three-body potential $v_3$ vanishes only at $\pi/2, \pi$, and $3\pi/2$, it is symmetric with respect to $\pi$, convex in $I_1$, and satisfies the following non-degeneracy and symmetry conditions:

$$v_3(\theta) > 8 \text{ if } \theta \in (\theta_{\min}, 2\pi/5],
\hspace{1cm} v_3(\theta) > 4 \text{ if } \theta \notin I,
\hspace{1cm} v_3(\theta) = v_3(\theta + \pi/2) = v_3(\theta + \pi) \text{ if } \theta \in I_1,$$

(4a)

and

$$v_3^{-1}(\frac{\pi}{2}) := \lim_{t \to 0} \frac{1}{t} v_3(t + \pi/2) < -\frac{2}{\pi}.$$

(4b)

We say that two particles $x_i$ and $x_j$ are bonded or that there is an (active) bond between $x_i$ and $x_j$, if $1 \leq \ell_{ij} < \ell^*$, in this case $v_2(\ell_{ij})$ is negative. The set $\mathcal{A}$ in (1) is defined as the set of all triples $(i, j, k)$ for which the angle $\theta_{ijk}$ separates two active bonds. The angle $\theta_{ijk}$ is said to be an (active) bond angle if $(i, j, k) \in \mathcal{A}$. We observe that the hard-interaction assumption $v_2 = \infty$ on $[0, 1]$ can be relaxed by asking $v_2$ to be very large in a right neighborhood of 0 (see [1, 3, 4]). The bond graph of a configuration $C_n$ is the graph consisting of all its vertices and active bonds. We note that the simple cycles of a bond graph are polygons, and that, since $v_2(\ell)$ vanishes for $\ell \geq \sqrt{2}$, every bond graph is a planar graph. Moreover, from (2) it follows that the minimal angle between two active bonds is $\theta_{\min}$ for all finite-energy configurations.

We say that a configuration is square if it is a (rotated and translated) subset of the square lattice $\mathbb{Z}^2$ (notice that the energy is invariant under rotations and translations). Furthermore, a configuration is connected if every particle is connected to every other by a simple path in the bond graph, and it is regular if every particle in it has at most four bonds, every bond angle is in $I$, and every simple cycle of its bond graph has at least four edges. Straightforward comparison arguments (competitors being constructed by simply moving a single particle) show that grounds states are regular, and that $E(C_n) \geq -b(C_n)$ (with equality corresponding to square configurations $C_n$) where $b(C_n)$ denotes the number of bonds in the bond graph of $C_n$.

Moreover, a connected configuration with $n \geq 4$ particles is said to be closed if it has no acyclic bonds, i.e., every bond is an edge of a simple cycle. Given a closed configuration $C_n$, we identify its boundary polygon consisting of (at least four) vertices and containing all the other particles in its interior region, and we denote it by $B(C_n)$. In addition, let $d(C_n)$ be the number of boundary particles, i.e. the particles of the boundary polygon, and $C_n^{\text{bulk}}$ be the bulk configuration consisting of the remaining $n - d(C_n)$ interior vertices. The bulk energy of $C_n$ is then defined by $E^{\text{bulk}}(C_n) := E(C_n^{\text{bulk}})$ and the boundary energy by $E^{\text{bound}}(C_n) := E(C_n) - E^{\text{bulk}}(C_n) = E(C_n) - E(C_n^{\text{bulk}})$. The set of all bonds and the set of all bond angles which are deactivated in $C_n$ by removing boundary particles are denoted, respectively, by $\Gamma(C_n)$ and by $\Theta(C_n)$. Since $v_2 \geq -1$, we obtain that the elementary inequality

$$E^{\text{bound}}(C_n) \geq -\#\Gamma(C_n) + \sum_{\theta_i \in \Theta(C_n)} v_3(\theta_i),$$

(5)

which reduces to $E^{\text{bound}}(C_n) = -\#\Gamma(C_n)$ in case of a square configuration $C_n$. 

3 The Square Crystallization Result

In this section we establish Theorem 1.1. We begin by constructing a ground-state candidate with $n \in \mathbb{N}$ particles that we denote by $D_n$. If $n = m^2$ for some $m \in \mathbb{N}$, we let $D_n$ be the $m \times m$ square in $\mathbb{Z}^2$, while if $n = m^2 + k$ for some $1 \leq k < 2m + 1$, we obtain $D_n$ by progressively adding the $k$ particles to $D_{m^2}$ at specific sites of $\mathbb{Z}^2$. In fact, we add the first particle right above the upper left corner of the $m \times m$ square, and then, if necessary, we clockwise add particles in such a way that each new particle is bonded to the previous one and, whenever possible, to the original $m \times m$ square. Notice that
b(D_n) may be computed by recursion: \( b(D_1) = 0, b(D_{n+1}) - b(D_n) = 1 \) if \( n = m^2 \) or \( n = m^2 + m \) for some \( m \in \mathbb{N} \), or \( b(D_{n+1}) - b(D_n) = 2 \) otherwise. It is not difficult to show that \( \beta(n) := [2n - 2\sqrt{n}] \) solves the recursion (see [3, Proposition 4.1]), so that indeed \( E(D_n) = -\beta(n) \).

We now observe that \( \#I(C_n) \) can be estimated in terms of \( d(C_n) \) for a regular and closed configuration \( C_n \). Let us denote by \( \varphi_i \) for \( i = 1, \ldots, ed \), the internal angles of \( B(C_n) \) that are in \( I_1 \), by \( \psi_i \), \( i = 1, \ldots, nd \), the ones in \( I_2 \) and by \( \xi_i \), \( i = 1, \ldots, vd \), the ones in \( I_3 \). Here, \( \varepsilon, \eta, \) and \( \nu \) are the ratios of the internal angles of \( B(C_n) \) that belong to \( I_1, I_2, \) and \( I_3 \) respectively. Since \( \sigma < \pi/8 \) in (3), given a boundary vertex \( x \) and the corresponding internal angles \( \theta \) of \( B(C_n) \), we infer that if \( \theta \) is in \( I_1 \), then \( x \) needs to be two-bonded, because otherwise there would be a bond angle at \( x \) smaller than \( \pi/16 \) and so not in \( I \). By a similar argument, if \( \theta \in I_2 \), then \( x \) is at most three-bonded, and if \( \theta \) is in \( I_3 \), then \( x \) has at most four bonds. It follows that

\[
\#I(C_n) \leq (1 + \varepsilon + 2\nu) d(C_n) = (\varepsilon + 2\eta + 3\nu) d(C_n). \tag{6}
\]

**Proof of Theorem 1.1. Step 1 (boundary energy estimate).** We here establish that \( E^{\text{bound}}(C_n) \geq -2d(C_n) + 4 \) holds for a regular and closed configuration \( C_n \), and that the inequality is strict in case of a non-square configuration \( C_n \) with a square \( C_n^{\text{bulk}} \). For simplicity we often omit the dependence on \( C_n \) in the formulas.

Since the sum of the internal angles of a polygon with \( d \) sides is \( \pi(d-2) \), we first observe that

\[
\varepsilon d\varphi + \eta d\psi + \nu d\xi = \pi(d-2), \quad \text{where} \quad \varphi := \frac{1}{ed} \sum_{i=1}^{ed} \varphi_i, \quad \psi := \frac{1}{nd} \sum_{i=1}^{nd} \psi_i, \quad \xi := \frac{1}{vd} \sum_{i=1}^{vd} \xi_i. \tag{7}
\]

By (4a) and the convexity of \( v_3 \) in \( I_1 \) we have \( v_3(\psi_i) \geq 2v_3(\psi_i/2) \) for \( i = 1, \ldots, nd \) and \( v_3(\xi_i) \geq 3v_3(\xi_i/3) \) for \( i = 1, \ldots, vd \). Still the convexity of \( v_3 \) in \( I_1 \), together with (5), (14) and (7), entails

\[
E^{\text{bound}} \geq -\varepsilon d - 2\nu d - 3\nu d + \sum_{i=1}^{ed} v_3(\varphi_i) + 2 \sum_{i=1}^{nd} v_3 \left( \frac{\psi_i}{2} \right) + 3 \sum_{i=1}^{vd} v_3 \left( \frac{\xi_i}{3} \right) \geq -\delta d + \delta dv_3(\alpha(\delta)), \tag{8}
\]

where \( \delta := \varepsilon + 2\eta + 3\nu \) and \( \alpha(\delta) := \pi - (d-2)/\delta \). By looking at (8), we immediately have that \( E^{\text{bound}}(C_n) \geq -2d(C_n) + 4 \) if \( \delta \leq \delta^* := 2 - 4/d \), otherwise, the same follows from the crucial hypothesis (4b) since (4b) implies

\[
v_3(\alpha(\delta)) \geq v_3 \left( \frac{\pi}{2} \right) + v_3 \left( \frac{\pi}{2} - \frac{\pi}{2} \right) \left( \alpha(\delta) - \frac{\pi}{2} \right) > -\frac{2}{\pi} \left( \alpha(\delta) - \frac{\pi}{2} \right) = \frac{\delta d - 2d + 4}{\delta d}. \tag{9}
\]

We now verify that if a bond in \( I_1 \) is not length 1 or a bond angle of \( \Theta \) is not in \( \{\pi/2, \pi, 3\pi/2\} \), then \( E^{\text{bound}} > -2d + 4 \). Notice that (8) has to hold with equality in order to have \( E^{\text{bound}} = -2d + 4 \). However, recalling (5), (8) is strict if the length of a bond in \( I_1 \) is not 1 or if an angle in \( \Theta \) which is adjacent to an interior vertex differs from \( \pi/2, \pi, or 3\pi/2 \). Otherwise, equality in (8) implies that \( \sum_{\theta \in \Theta} v_3(\theta) = \delta dv_3(\alpha(\delta)) \), but this relation, taking the strict inequality in (9) into account, readily entails \( \delta = \delta^* \), thus \( \alpha(\delta) = \pi/2 \), and this shows that all the other angles of \( \Theta \) are in \( \{\pi/2, \pi, 3\pi/2\} \) as well.

**Step 2 (induction on bond graph layers).** In this step we follow the classical induction argument introduced by Rabin in [5] that has been recently revisited in [3, 4]. We refer the reader to [3, 4] for more details. We begin by observing that the statement of the Theorem is trivial for \( n \leq 4 \), and we then proceed by induction. We prove that if the assertion holds for a ground state \( C_m \) with \( m < n \), then it holds also for \( C_n \). By contradiction we suppose that \( C_n \) is not square and we observe that a contradiction may be easily reached for a configuration \( C_n \) that is not closed by considering subconfigurations and by using elementary properties of the function \( \beta(n) \) (see [3]). Instead, for a closed configuration \( C_n \) that is not square, we argue as follows. Either \( C_n^{\text{bulk}} \) is not square itself and hence, by induction we have that \( E^{\text{bulk}} > -\beta(n - d) \), or \( C_n^{\text{bulk}} \) is square and so from Step 1 it follows that \( E^{\text{bound}} > -2d + 4 \). Therefore, by Step 1, in both cases we obtain that

\[
E = E^{\text{bulk}} + E^{\text{bound}} > -2(n-d) - 2\sqrt{n-d} - 2d + 4. \tag{10}
\]

Finally, by combining (10) with the Euler formula for planar graphs we have that \( E > -\beta(n) \) holds (see [3]), and this contradicts the fact that \( C_n \) is a ground state.

\[\square\]

## 4 Isoperimetric Inequality and Convergence to the Wulff Shape

Let us define the area \( A \) and the perimeter \( P \) of a regular configuration \( C_n \) by \( A(C_n) := L^2(F(C_n)) \) and \( P(C_n) := \mathcal{H}^1(F(C_n)) + 2\mathcal{H}^1(G(C_n)) \), respectively, where \( F(C_n) \subset \mathbb{R}^2 \) is the closure of the union of the regions enclosed by the simple cycles of \( C_n \) that consists of only 4 bonds, \( G(C_n) \subset \mathbb{R}^2 \) is the union of all bonds which are not included in \( F(C_n) \), and \( \mathcal{H}^1 \) denotes the one-dimensional Hausdorff measure. For square configurations, we easily see that \( E(C_n) = -2A(C_n) - \frac{1}{2}P(C_n) \).

We characterize ground states as solutions of a discrete isoperimetric problem. In the following, \([x] := \min\{z \in \mathbb{Z} : x \leq z\} \).
Theorem 4.1  Every connected square configuration \( C_n \) with \( n > 1 \) satisfies
\[
\sqrt{\lambda(C_n)} \leq k_n P(C_n) \quad \text{where} \quad k_n := \frac{\sqrt{n - 2\sqrt{n-1}}}{2[2\sqrt{n-1} - 2].} \tag{11}
\]
Moreover, ground states correspond to those configurations for which (11) holds with the equality, and, equivalently, to those configurations that attain the maximum area, i.e. \( n - 2\sqrt{n-1} \), and the minimum perimeter, i.e. \( 2[2\sqrt{n-1} - 2]. \)

Proof. Step 1. We begin by establishing that (11) holds for a particular class of configurations called quasirectangles and denoted by \( R^{r,c,e}_n \), where the triplet \((r, c, e)\) is assumed to be in \( T_n := \{(r, c, e) \in \mathbb{N}^3 \mid r \leq c, 1 \leq e \leq c, re + e = n\}. \) A quasirectangle \( R^{r,c,e}_n \) is a square configuration obtained by adding a connected line of \( e \) extra particles to the \((r \times c)\)-rectangle (consisting of \( r \) aligned rows each with \( c \) particles) in such a way that each extra particle is bonded to one and only one particle of the rectangle. Let us define \( k_n \) by
\[
\begin{align*}
k_n := \max_{(r,c,e) \in T_n} \frac{\sqrt{\lambda(R^{r,c,e}_n)}}{P(R^{r,c,e}_n)} &= \max \left\{ \frac{\sqrt{n - (r+c)}}{2(r+c) - 2} \mid r, c \in \mathbb{N} \text{ and } n - \max \{r, c\} \leq rc < n \right\}.
\end{align*}
\tag{12}
\]
where the second equality follows from the fact that \( \lambda(R^{r,c,e}_n) = (r-1)(c-1) + e - 1 = n - (r+c) \) and \( P(R^{r,c,e}_n) = 2(r+c) - 2. \) Thus, \( k_n \) is realized at the minimum admissible value of \( r+c \) that can be analytically computed to be equal to \( \ell_n := \lceil 2\sqrt{n-1} \rceil. \)

Observe that the configuration \( D_n \) introduced in Section 3 is a quasirectangle that realizes the minimum in (12) since the sum of its rows and columns is exactly \( \ell_n. \) Therefore, we have that \( \sqrt{\lambda(D_n)} = k_n P(D_n), \) and from the same reasoning it follows also that the maximum area and the minimum perimeter among quasirectangles are realized by \( \lambda(D_n) = n - \ell_n \) and \( P(D_n) = 2\ell_n - 2, \) respectively. Inequality (11) is now a direct consequence of the fact that we can always rearrange the particles of a connected square configuration \( C_n \) in a quasirectangle without increasing its perimeter and decreasing its area, see [3, Lemma 7.3]. Therefore, it follows also that \( \lambda(D_n) \) maximizes the area and \( P(D_n) \) minimizes the perimeter among all connected square configurations.

Step 2. We now prove the second statement. Every connected square configuration \( C_n \) that satisfies \( \sqrt{\lambda(C_n)} = k_n P(C_n) \) is a ground state since by the elementary relation \( E(C_n) = -2\lambda(C_n) - \frac{1}{2}P(C_n) \) we have that
\[
E(D_n) \leq E(C_n) = -2\lambda(C_n) - \frac{P(C_n)}{2} = -2k_n^2 P^2(C_n) - \frac{P(C_n)}{2} \leq -2k_n^2 P^2(D_n) - \frac{P(D_n)}{2} = E(D_n),
\]
where we used the fact that \( D_n \) is a ground state that minimizes the perimeter as established in Step 1.

Proof of Theorem 1.2. Let \( C_n \) be a ground state and \( R(C_n) \) be the minimal \( (\ell_1 - 1) \times (\ell_2 - 1) \)-rectangle (with \( \ell_1 \geq \ell_2 \) and sides parallel to the two directions of its unit square lattice of reference, say \( \mathbb{L} \)) that contains \( C_n. \) Since every ground state \( C_n \) is connected in the directions of the square lattice, by Theorem 4.1 we obtain that \( \ell_1 + \ell_2 = \frac{1}{2} P(C_n) + 2 = 2\lceil \sqrt{n-1} \rceil + 1 = \ell_n + 1. \) By solving the maximum problem \( \ell_1 - \ell_2 := \max \{a - b : a, b \in \mathbb{N}, ab \geq n, a + b = \ell_n + 1\} \) we observe that
\[
\ell_1 - \ell_2 = 2 \left[ \frac{\sqrt{\sqrt{n} + \sqrt{(\sqrt{n})^2 - 4n}}}{2} - \frac{\sqrt{2\sqrt{n}-1}}{2} \right] \leq 2n^{1/4} + 1.
\tag{13}
\]
and hence, we have \( \sqrt{n} \leq \ell_1 \leq \sqrt{n} + n^{1/4} + 1 \) and \( \sqrt{n} - n^{1/4} - 1 \leq \ell_2 \leq \sqrt{n}. \) Therefore, we obtain that \( d(R'_n, S_n) \leq n^{1/4} + 2 \) where \( d \) denotes the Hausdorff distance and \( R'_n \) is a suitable rotation and translation of \( R_n := R(C_n) \cap \mathbb{L}. \) Let us now define \( C'_n \) as the analogous rotation and translation of \( C_n. \) Since we have that
\[
\#(C'_n \Delta S_n) \leq \#(R'_n \setminus S_n) + \#(S_n \setminus R'_n) + \#((R'_n \cap S_n) \setminus C'_n)
\]
\[
\leq 2\#(R'_n \setminus S_n) + \#(S_n \setminus R'_n) \leq 2(\ell_2 + \sqrt{n}) d(R'_n, S_n) \leq (3\sqrt{n} + 1)(n^{1/4} + 2),
\tag{14}
\]
the assertion follows.

References