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Abstract

We give a sufficient condition on a radial parametrized long-range potential for a compact local minimality of a given d-dimensional Bravais lattice for its total energy of interaction. This work is widely inspired by the paper of F. Theil about two dimensional crystallization.

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1 Introduction

As explained in [2], the crystallization problem, that is to say to understand why the particles structures are periodic at low temperature, is difficult and still open in the main cases. Theil exhibited in [7] a radial parametrized long-range potential with the same form as the Lennard-Jones potential such that the triangular lattice is the ground state of the total energy in the sense of thermodynamic limit. This kind of potential, parametrized by a real $\alpha > 0$, is bigger than α^{-1} close to the origin, corresponding to Pauli's principle, has a well centred in 1 and a 2α width, its second derivative at 1 is strictly positive and its decay at infinity is $r \mapsto \alpha r^{-7}$. Thus, as small is α, as close to 1 is the mutual distance between nearest neighbours of the ground state configuration and as the interactions between distant points are negligible.

In this paper, our idea is to present a parametrized potential very close to this one, with the most natural possible assumptions, such that a given Bravais lattice L of $\mathbb{R}^d$ is a N-compact local minimum for the total energy of interaction. This kind of local minimality is called "N-compact" because, given a maximal number N of points that we want to move a little bit, there exists a maximal perturbation of the points which gives a larger total energy of interaction, in the sense of the difference of energies is positive. Moreover, as small is the parameter, as large the number N can be taken. We strongly inspire Theil's potential, keeping only local assumptions and strong parametrized decay. Furthermore, our work can be related to that of Torquato et al. about targeted self-assembly [5, 8] where they search radial potentials such that a given configuration – more precisely a part of a lattice – is a ground state for the total energy of interaction.

After defining the concepts and our parametrized potentials, we give the theorem, its proof and some important remarks and applications.

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2 Preliminaries : Bravais lattice and N-compact local minimality

Definition 2.1. *Let $d \in \mathbb{N}^*$, $(u_1, ..., u_d)$ be a basis of $\mathbb{R}^d$ and $L = \bigoplus_{i=1}^n u_i \subset \mathbb{R}^d$ be a Bravais lattice. For any $\lambda > 0$, we define $m(\lambda) := \sharp\{L \cap \{\|x\| = \lambda\}\}$ where $\|.\|$ denote the Euclidean norm and $\sharp A$ is the cardinal of set A. Moreover, we call $\lambda_1 := \min\{\|x\|; x \in L^*\}$, where $L^* = L\backslash\{0\}$ and $\lambda_2 := \min\{\|x\|; \|x\| > \lambda_1, x \in L\}$.*
Furthermore, for a Bravais lattice $L \subset \mathbb{R}^d$, we define the following both lattice sums, for $n > d$,

$$\zeta_L^*(n) := \sum_{\substack{x \in L \\ \|x\| > \lambda_1}} \|x\|^{-n},$$
$$\bar{\zeta}_L(n) := \sum_{\substack{x \in L \\ \|x\| > \lambda_1}} (\|x\| - \lambda_1)^{-n}.$$

Definition 2.2. *Let $L \subset \mathbb{R}^d$ be a Bravais lattice, $B \subset L$ a finite subset and α be a real such that $0 < \alpha < \lambda_1/2$. We say that B^α is an α-compact perturbation of B if*

$$\forall b \in B, \exists! b^\alpha \in B^\alpha \text{ such that } \|b - b^\alpha\| \leq \alpha.$$

Moreover, if B^α is an α-perturbation of $B \subset L$, we write $L^\alpha(B) := (L\backslash B) \cup B^\alpha$ the perturbed lattice.

Definition 2.3. *Let $d \in \mathbb{N}^*$. We say that $V : \mathbb{R}_+^* \to \mathbb{R}$ is a **d-admissible potential** if V is a C^3 function and, for any Bravais lattice $L \subset \mathbb{R}^d$,*

$$\sum_{x \in L^*} |V(\|x\|)| + \sum_{x \in L^*} \|x\||V'(\|x\|)| + \sum_{x \in L^*} \|x\|^2|V''(\|x\|)| + \sum_{x \in L^*} \|x\|^3|V'''(\|x\|)| < +\infty.$$

Remark 2.1. If, for any $k \in \{0, 1, 2, 3\}$, $|V^{(k)}(r)| = O(r^{-p_k})$, $p_k > d$, then V is d-admissible.

Definition 2.4. *Let L be a Bravais lattice of $\mathbb{R}^d$ and V a d-admissible potential. Let $N \in \mathbb{N}^*$, we say that L is a **N-compact local minimum for the total V-energy** if for any subset $B \subset L$ such that $\sharp B \leq N$, there exists $\alpha_0 > 0$ such that for any $\alpha \in [0, \alpha_0)$ and any α-compact perturbation B^α of B,*

$$\Delta_L^\alpha(V; B) := \sum_{b^\alpha \in B^\alpha} \sum_{\substack{y \in L^\alpha(B) \\ y \neq b^\alpha}} V(\|b^\alpha - y\|) - \sum_{b \in B} \sum_{\substack{x \in L \\ x \neq b}} V(\|b - x\|) \geq 0.$$

3 Parametrized potential and main result

Definition 3.1. *Let $L \subset \mathbb{R}^d$ be a Bravais lattice. We call **parametrized L-potential** every d-admissible function $V_\theta : \mathbb{R}_+^* \to \mathbb{R}$, defined for fixed $\theta \in [0, \lambda_1/2)$, satisfying :*

1. ***Zero pressure condition :*** *it holds $\sum_{x \in L^*} \|x\| V_\theta'(\|x\|) = 0$;*
2. ***Parametrized fast decay :*** *$\exists r_0 \in [\lambda_1, \lambda_2)$, $\exists \varepsilon > 0$, $\exists p > 3$ such that for any $r > r_0$, $|V_\theta''(r)| \leq \theta^{1+\varepsilon} r^{-p-2}$;*
3. ***Local convexity around first neighbours :*** *$V_\theta''(\lambda_1)$ is independent of θ and $V_\theta''(\lambda_1) > 0$;*
4. ***Bounded third derivative :*** *there exists $M > 0$, independent of θ, such that, for any $\lambda_1/2 < r < \lambda_2$, $|V_\theta'''(r)| \leq M$.*

THEOREM 3.1. *Let $L \subset \mathbb{R}^d$ be a Bravais lattice, then for any $N \in \mathbb{N}^*$, there exists $\theta_0 > 0$ such that for every $\theta \in [0, \theta_0]$ and every parametrized L-potential V_θ, L is a N-compact local minimum for the total V_θ-energy. Furthermore, in this case, the maximal perturbation α_0 can be chosen equal to θ.*

Proof. Let L be a Bravais lattice of $\mathbb{R}^d$. Let $N \in \mathbb{N}^*$ and $B := \{b_1, ..., b_N\}$. Let α_0 be such that $0 \leq \alpha_0 < \lambda_1/2$ and $B^{\alpha_0} = \{b_1^{\alpha_0}, ..., b_N^{\alpha_0}\}$ a α_0-compact perturbation of B. For any $1 \leq i \leq N$, for any $y \in L^{\alpha_0}(B)$, $y \neq b_i^{\alpha_0}$, and $x \in L$ such that $\|x - y\| \leq \alpha_0$, we define

$$\alpha_{i,x} := \|b_i^{\alpha_0} - y\| - \|b_i - x\|.$$

We assume, without loss of generality, that $\max_{i,x} |\alpha_{i,x}| = 2\alpha_0$, left to decrease α_0. We set $\theta \in [0, \lambda_1/2)$ and V_θ a L-parametrized potential. We have

$$\Delta_L^{\alpha_0}(V_\theta; B) = \sum_{i=1}^{N} \sum_{\substack{y \in L^{\alpha_0}(B) \\ y \neq b_i^{\alpha_0}}} V_\theta(\|b_i^{\alpha_0} - y\|) - \sum_{i=1}^{N} \sum_{\substack{x \in L \\ x \neq b_i}} V_\theta(\|b_i - x\|).$$

By Taylor expansion, we get, for any $1 \leq i \leq N$, for any $y \in L^{\alpha_0}(B)$, $y \neq b_i^{\alpha_0}$, and $x \in L$ such that $\|x - y\| \leq \alpha_0$,

$$V_\theta(\|b_i^{\alpha_0} - y\|) \geq V_\theta(\|b_i - x\|) + \alpha_{i,x} V_\theta'(\|b_i - x\|) + \frac{\alpha_{i,x}^2}{2} V_\theta''(\|b_i - x\|) - \frac{|\alpha_{i,x}|^3}{6} \|V_\theta'''\|_{i,x}$$

where $\|V_\theta'''\|_{i,x} := \sup\left\{|V_\theta'''(r)|; \|b_i - x\| - \alpha_{i,x} < r < \|b_i - x\| + \alpha_{i,x}\right\}$. Hence we obtain

$$\Delta_L^{\alpha_0}(V_\theta; B) \geq \sum_{i=1}^{N} \sum_{\substack{x \in L \\ x \neq b_i}} \alpha_{i,x} V_\theta'(\|b_i - x\|) + \frac{1}{2} \sum_{i=1}^{N} \sum_{\substack{x \in L \\ x \neq b_i}} \alpha_{i,x}^2 V_\theta''(\|b_i - x\|) - \frac{1}{6} \sum_{i=1}^{N} \sum_{\substack{x \in L \\ x \neq b_i}} |\alpha_{i,x}|^3 \|V_\theta'''\|_{i,x}.$$

Now we cut interactions into two parts : the short range and the long range. For any $1 \leq i \leq N$, we set

$$\mathcal{S}_L^i := \{x \in L; \|x - b_i\| = \lambda_1\} \quad \text{and} \quad \mathcal{L}_L^i := \{x \in L; \|x - b_i\| > \lambda_1\}.$$

As we assume that for all $r > r_0$ $|V_\theta'''(r)| \leq \theta^{1+\varepsilon} r^{-p-2}$ and V_θ',V_θ'' go to 0 at $+\infty$, we have, by a simple argument, that $|V_\theta'(r)| \leq \theta^{1+\varepsilon} r^{-p}$ and $|V_\theta''(r)| \leq \theta^{1+\varepsilon} r^{-p-1}$ for all $r > r_0$, therefore we get :

$$\sum_{i=1}^{N} \sum_{\substack{x \in L \\ x \neq b_i}} \alpha_{i,x} V_\theta'(\|b_i - x\|) \geq V_\theta'(\lambda_1) \left(\sum_{i=1}^{N} \sum_{x \in \mathcal{S}_L^i} \alpha_{i,x} \right) - 2\alpha_0 \theta^{1+\varepsilon} \sum_{i=1}^{N} \sum_{x \in \mathcal{L}_L^i} \frac{1}{\|b_i - x\|^p},$$

$$\frac{1}{2} \sum_{i=1}^{N} \sum_{\substack{x \in L \\ x \neq b_i}} \alpha_{i,x}^2 V_\theta''(\|b_i - x\|) \geq \frac{V_\theta''(\lambda_1)}{2} \left(\sum_{i=1}^{N} \sum_{x \in \mathcal{S}_L^i} \alpha_{i,x}^2 \right) - 2\alpha_0^2 \theta^{1+\varepsilon} \sum_{i=1}^{N} \sum_{x \in \mathcal{L}_L^i} \frac{1}{\|b_i - x\|^{p+1}}.$$

As $\sum_{x \in L^*} \|x\| V_\theta'(\|x\|) = 0$, we have $V_\theta'(\lambda_1) = -\frac{1}{m(\lambda_1)\lambda_1} \sum_{\substack{x \in L \\ |x| > \lambda_1}} \|x\| V_\theta'(\|x\|)$. As L is a Bravais lattice, for any $1 \leq i \leq N$, $\sharp \mathcal{S}_L^i = m(\lambda_1)$ and we obtain

$$\left| V_\theta'(\lambda_1) \left(\sum_{i=1}^{N} \sum_{x \in \mathcal{S}_L^i} \alpha_{i,x} \right) \right| \leq \frac{1}{m(\lambda_1)\lambda_1} \left(\sum_{x \in \mathcal{L}_L^0} \|x\| |V_\theta'(\|x\|)| \right) \left(\sum_{i=1}^{N} \sum_{x \in \mathcal{S}_L^i} |\alpha_{i,x}| \right) \leq \frac{2\alpha_0 \theta^{1+\varepsilon} N}{\lambda_1} \zeta_L^*(p-1).$$

As L is a Bravais lattice, we have, for any $b_i \in B$ and any $p > d$, $\displaystyle\sum_{x\in\mathcal{L}_L^i} \frac{1}{\|b_i - x\|^p} = \zeta_L^*(p)$, hence

$$\sum_{i=1}^{N} \sum_{\substack{x\in L\\ x\neq b_i}} \alpha_{i,x} V_\theta'(\|b_i - x\|) \geq -2\alpha_0\theta^{1+\varepsilon} N \left(\lambda_1^{-1}\zeta_L^*(p-1) + \zeta_L^*(p)\right).$$

As $\max\limits_{i,x} |\alpha_{i,x}| = 2\alpha_0$, we have $\displaystyle\sum_{i=1}^{N}\sum_{x\in\mathcal{S}_L^i} \alpha_{i,x}^2 \geq 4\alpha_0^2$, and we obtain

$$\frac{1}{2}\sum_{i=1}^{N} \sum_{\substack{x\in L\\ x\neq b_i}} \alpha_{i,x}^2 V_\theta''(\|b_i - x\|) \geq 2V_\theta''(\lambda_1)\alpha_0^2 - 2N\alpha_0^2\theta^{1+\varepsilon}\zeta_L^*(p+1).$$

Now we remark that $\displaystyle -\frac{1}{6}\sum_{i=1}^{N} \sum_{\substack{x\in L\\ x\neq b_i}} |\alpha_{i,x}|^3 \|V_\theta'''\|_{i,x} \geq -\frac{4}{3}\alpha_0^3 \sum_{i=1}^{N} \sum_{\substack{x\in L\\ x\neq b_i}} \|V_\theta'''\|_{i,x}$. Moreover,

$$\begin{aligned}
\sum_{i=1}^{N} \sum_{\substack{x\in L\\ x\neq b_i}} \|V_\theta'''\|_{i,x} &= \sum_{i=1}^{N}\sum_{x\in\mathcal{S}_L^i} \|V_\theta'''\|_{i,x} + \sum_{i=1}^{N}\sum_{x\in\mathcal{L}_L^i} \|V_\theta'''\|_{i,x} \\
&\leq MNm(\lambda_1) + \theta^{1+\varepsilon}\sum_{i=1}^{N}\sum_{x\in\mathcal{L}_L^i} \frac{1}{(\|b_i - x\| - \alpha_{i,x})^{p+2}} \\
&\leq MNm(\lambda_1) + \theta^{1+\varepsilon} N\bar{\zeta}_L(p+2),
\end{aligned}$$

because $\alpha_{i,x} < \lambda_1$. Finally we get, for any $0 \leq \alpha_0 < \lambda_1/2$,

$$\begin{aligned}
\Delta_L^{\alpha_0}(V_\theta; B) \geq & 2V_\theta''(\lambda_1)\alpha_0^2 - 2N\left[\alpha_0^2\theta^{1+\varepsilon}\zeta_L^*(p+1) + \frac{2}{3}M\alpha_0^3 m(\lambda_1) + \frac{2}{3}\alpha_0^3\theta^{1+\varepsilon}\bar{\zeta}_L(p+2)\right. \\
& \left. + \alpha_0\theta^{1+\varepsilon}\left(\lambda_1^{-1}\zeta_L^*(p-1) + \zeta_L^*(p)\right)\right].
\end{aligned}$$

Given $\theta \in [0, \lambda_1/2)$, if $\alpha_0 = \theta$, then there exist positive real A, B, C, D, independent of θ, such that

$$\Delta_L^{\theta}(V_\theta; B) \geq 2V_\theta''(\lambda_1)\theta^2 - N\left(A\theta^{2+\varepsilon} + B\theta^3 + C\theta^{3+\varepsilon} + D\theta^{4+\varepsilon}\right). \tag{3.1}$$

As $V_\theta''(\lambda_1) > 0$ is also independent of θ, there exists $\theta_0 \in [0, \lambda_1/2)$, depending on N, sufficiently small such that for any $\theta \in [0, \theta_0]$ and for any $\alpha \in [0, \theta]$, $\Delta_L^{\alpha}(V_\theta; B) \geq 0$ and then L is a N-compact local minimum for the total V_θ-energy, for any parametrized L-potential V_θ. □

4 Remarks

1. Zero pressure condition and local minimality among dilated of L. As explained for instance in [1], if $E_{V_\theta}[L] := \displaystyle\sum_{x\in L^*} V_\theta(\|x\|)$ is the energy per particle of L, i.e. the free energy at zero temperature, then, by usual thermodynamics formula, we define pressure P and isothermal compressibility κ_T by

$$P = -\frac{dE_{V_\theta}[L]}{dA} = -\frac{1}{2A}\sum_{x\in L^*} \|x\| V_\theta'(\|x\|),$$

$$\frac{1}{\kappa_T} = -A\frac{dP}{dA} = A\frac{d^2E_{V_\theta}[L]}{dA^2} = \frac{1}{4A}\sum_{x\in L^*}\left[\|x\|^2V_\theta''(\|x\|) - \|x\|V_\theta'(\|x\|)\right].$$

where A is the area per particle. As we want to have L as a N-compact local minimum for arbitrary large N in an infinite volume, it is thermodynamically natural – for instance if L is the cooling of an ideal gas – to suppose $P = 0$ at zero temperature, which gives a kind of justification of the necessity of the zero pressure condition 1. in definition 3.1.

Moreover, we know that $\kappa_T > 0$ (see [6, Section 5.1]), therefore $\sum_{x\in L^*}\|x\|^2V_\theta''(\|x\|) > 0$. Actually, that follows here from assumptions on V_θ, if L is a N-compact local minimum for the total V_θ-energy with N sufficiently large. Indeed, by assumption, we have, for θ sufficiently small,

$$\sum_{x\in L^*}\|x\|^2V_\theta''(\|x\|) \geq m(\lambda_1)\lambda_1V_\theta''(\lambda_1) - \theta^{1+\varepsilon}\zeta_L^*(p+1) > 0.$$

Now if we consider $f : r \mapsto E_{per}[V_\theta; rL]$, we get, by $P = 0$ and $\kappa_T \geq 0$, $f'(1) = 0$ and $f''(1) > 0$ and L **is a local minimum of** $L \mapsto E_{per}[V_\theta; L]$ **among dilated of** L, which seems natural if L is a N-compact local minimum for the total V_θ-energy for N sufficiently large., which is actually assumed in Theil's paper [7].

However, the reverse is false. A Bravais lattice can be a local minimum among its dilated for the energy per point but not a N-compact local minimum for the total energy. For instance, if $d = 1$, $L = \mathbb{Z}$, $N = 1$ and V defined by $V(r) = 0$ for $r \geq 5/2$, $V'(1) = V'(2) = 0$, $V''(1) = -1$ and $V''(2) = 1/3$. We have $\sum_{x\in\mathbb{Z}^*}|x|V'(|x|) = 0$ and $\sum_{x\in\mathbb{Z}^*}|x|^2V''(|x|) = 2/3 \geq 0$ hence $\mathbb{Z}$ is a local minimum among lattices of the V-energy per point. For $\alpha \geq 0$, we estimate, by Taylor expansion,

$$\begin{aligned}\Delta^\alpha(V;L) &= \sum_{x\in\mathbb{Z}^*}\left[V(|x-\alpha|) - V(|x|)\right]\\ &= V(1-\alpha) + V(1+\alpha) - 2V(1) + V(2-\alpha) + V(2+\alpha) - 2V(2)\\ &= \alpha^2V''(1) + \alpha^2V''(2) + \alpha^2\phi(\alpha) = \alpha^2(-2/3 + \phi(\alpha))\end{aligned}$$

where $\phi(\alpha)$ goes to 0 as $\alpha \to 0$. Hence for $\alpha < \alpha_0$ sufficiently small, $-2/3 + \phi(\alpha) < 0$ and $\mathbb{Z}$ is not a 1-compact local minimum of the total V-energy.

2. Effects of parameters ε, p and $V_\theta''(\lambda_1)$. By (3.1), our assumptions on V_θ give indications about the stability of lattice L:

- Range : a larger p or a larger ε allow to take a larger perturbation α_0 for fixed N, i.e. a better "collapse" at infinity implies a stronger stability of the lattice;
- Second derivative around nearest-neighbours distance : a larger $V_\theta''(\lambda_1)$ also allows a larger perturbation α_0 for fixed N. Typically, a narrow well around λ_1 "catches" the first neighbours of the minimizing configuration at distance λ_1.

3. Difference between the collapse after the first distance and the perturbation. We can see that $\theta^{1+\varepsilon} << \theta$, i.e. the collapse is really smaller than the perturbation and this allows to do not assume a local behaviour of V_θ around λ_1 with respect to θ, as in Theil's work. Obviously, if $\theta = 0$ then $V_0(r) = 0$ for any $r > r_0$ and $V_0'(\lambda_1) = 0$, therefore λ_1 is a local minimum of V_0 and the potential is short-range : only the first neighbours are important and the N-compact local minimality is clear for any N with a perturbation α_0 as small as N is large.

4. A kind of Cauchy-Born rule. Our result can be viewed like a justification of a kind of Cauchy-Born rule (see [4, 3]). Indeed, if we consider a solid as a Bravais lattice L where the inside is a finite part of L with cardinal N and the rest is its boundary, a (small) linear perturbation of the inside, depending on N, increases the total energy of interaction in the solid. That is to say that the inside of the solid follows its boundary to a stable configuration.

5. Numerical example. If we let $d = 2$, $V_\theta''(1) = 1$, $M = 1$, $p = 4$; $\varepsilon = 1$ and L is the triangular lattice of length 1, i.e. $L = A_2 = \mathbb{Z}(1,0) \oplus \mathbb{Z}(1/2, \sqrt{3}/2)$, we get $\zeta_L^*(p-1) \approx 4.9616984$, $\zeta_L^*(p) \approx 1.710774$, $\zeta_L^*(p+1) \approx 0.761895$, $\bar{\zeta}_L(p+2) \approx 15.50957$. Hence, by (3.1), we have

$$\Delta_{A_2}^\theta(V_\theta; B) \geq 2\theta^2 \left[1 - N(10.3397\theta^3 + 0.761895\theta^2 + 10.67247\theta^3)\right],$$

and we find, for any $k \in \mathbb{N}$, if $N = 10^k$, then the maximal perturbation is a least $\theta_0 \approx 10^{-k-1}$ and the collapse is at least $\theta_0^{1+\varepsilon} \approx 10^{-2k-2}$.
Actually, (3.1) is true for any $\theta \in [0, \theta_0]$ if

$$\theta_0 \leq \left(\frac{V_\theta''(\lambda_1)}{\Phi}\right)^{1/\varepsilon} \times N^{-1/\varepsilon}$$

where $\Phi = \zeta_L^*(p+1) + \frac{2}{3}m(\lambda_1)M + \frac{2}{3}\bar{\zeta}_L(p+2) + \lambda_1^{-1}\zeta_L^*(p-1) + \zeta_L^*(p)$, which gives a computable lower bound of a maximal perturbation of a finite set with cardinal N.

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