Cauchy-Born strain energy density for coupled incommensurate elastic chains

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Abstract

The recent fabrication of weakly interacting incommensurate two-dimensional lattices [9] requires an extension of the classical notion of the Cauchy-Born strain energy density since these atomistic systems are not periodic. In this paper, we rigorously formulate and analyze a Cauchy-Born strain energy density for weakly interacting incommensurate one-dimensional lattices (chains) as a large body limit and we give error estimates for its approximation by the popular supercell method.

Introduction

Graphene has recently been isolated as a free-standing two-dimensional hexagonal lattice [4]. Its continuum mechanical deformation can be modeled by the classical Cauchy-Born strain energy density [2,3,18], which is defined as the atomistic potential energy density of the unit cell for a homogeneously deformed lattice or multilattice [8,15,17,21].

Even more recently, weakly interacting incommensurate two-dimensional lattices such as single layer molybdenum disulfide (MoS_2) on graphene have been fabricated with the potential for improved design of electronic properties [9]. The classical Cauchy-Born rule can be used to link the macroscopic mechanical deformations to atomic displacements in independent monolayers such as graphene since they are multilattices, see e.g. [1], but an extension of the classical Cauchy-Born energy density is needed to accurately model the deformation of few layers stackings of weakly interacting incommensurate two-dimensional lattices since these atomistic systems have no periodicity, either due to differences in lattice constants or to a rotation angle between the respective lattices.

The Cauchy-Born strain energy density has been used to coarse-grain the deformation of a lattice away from defects in hybrid atomistic-to-continuum methods [7,14–16]. The extension of these hybrid atomistic-to-continuum methods to incommensurate systems

introduces the additional errors analyzed in this paper since the incommensurate Cauchy-Born strain energy density must be approximated on supercells.

In this work, we consider some simple one-dimensional toy models as a setting in which to rigorously formulate and analyze a Cauchy-Born strain energy density for weakly interacting incommensurate two-dimensional lattices as a large body limit. In particular, we show that the convergence with respect to system size of the energy density of incommensurate system is slow and very nonuniform, but higher convergence rates can be obtained for numerical computations when the system size is the denominator of a rational approximation of the irrational (incommensurate) ratio of the lattice spacing of the two weakly interacting chains. We also give error estimates for the popular supercell approximation which is based on approximating an incommensurate multi–layer system by a sequence of periodic configurations [12, 20].

1 An elementary model

1.1 Finite system

Let us first study a simple problem presented in Figure 1.1. We consider two parallel one-dimensional chains of atoms of approximate length $L \geq 1$, characterized by their isolated ground state lattice constants, respectively 1 for chain $\mathcal{C}^1$ and α for chain $\mathcal{C}^\alpha$, where α is an irrational real number in $(0, 1)$. The chains are separated by a fixed distance. In each chain, atoms interact via a smooth nearest-neighbor atomic potential, respectively $\psi_1(\Delta s)$ and $\psi_\alpha(\Delta s)$ where Δs is the distance between the atoms.

The two chains are also interacting through a long-range smooth pairwise atomic potential, such as the Lennard-Jones or Morse potentials. This potential can be rewritten as $V_{int}(\Delta s)$ where Δs is the abscissa difference between two atoms of each chain along the direction of the chains, not their respective distance. The goal is to compute the elastic resistance of the coupled system when the left end at 0 is held fixed, while a force is applied at the right end at L.

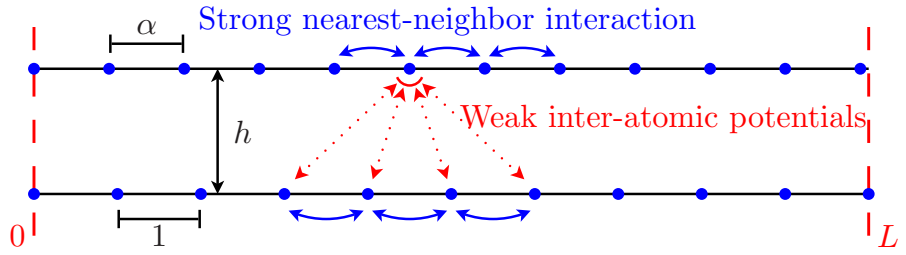


Figure 1: Sketch of a system of weakly coupled incommensurate atomic chains.

We assume that the intra-chain interaction (modeling covalent bonds) is much stronger than the inter-chain interaction (modeling Van-der-Waals forces). We thus suppose that the atoms in each chain are always equally spaced and the chains deform uniformly. When the applied force moves the right end of the system from L to $(1 + \varepsilon)L$, the atom

positions are given by:

$$\begin{cases} S_i^1 = (1+\varepsilon)i & \text{for } 0 \le i \le [L] \text{ for atoms in the first chain,} \\ S_j^\alpha = (1+\varepsilon)\alpha j & \text{for } 0 \le j \le [L/\alpha] \text{ for atoms in the second chain,} \end{cases} \tag{1.1}$$

where $[\cdot]$ denotes the closest integer to the real argument. Note that the lattice constants remain incommensurate in the deformed state since their ratio remains α. Thanks to this rigidity assumption, the rescaled potential energy of the coupled system per unit length is given exactly by:

$$\begin{aligned} \mathcal{E}(\varepsilon; L) = \frac{1}{L}\left([L]\psi_1(1+\varepsilon) + [\alpha^{-1}L]\psi_\alpha(\alpha+\varepsilon\alpha)\right) \\ + \frac{1}{L}\sum_{i=0}^{[L]}\sum_{j=0}^{[\alpha^{-1}L]} V_{int}\left((\alpha j - i)(1+\varepsilon)\right). \end{aligned} \tag{1.2}$$

Note that the elastic constant per unit length of the system can be deduced from (1.2) as

$$\mathcal{K}(\varepsilon_{eq}; L) = \frac{\partial^2 \mathcal{E}}{\partial \varepsilon^2}(\varepsilon_{eq}(L); L) \tag{1.3}$$

where $\varepsilon_{eq}(L)$ corresponds to a minimum of the function $\mathcal{E}(\varepsilon; L)$, i.e., characterizes a ground state of the coupled system.

1.2 Limit behavior.

Let us now study the elastic behavior of the system in the limit $L \to \infty$, and in particular the definition of a length-independent elastic constant as the limit of (1.3). We will assume that the long-range potential V_{int} is smooth and decays fast enough at infinity: for a given $\eta > 0$,

$$\begin{aligned} V_{int}(s) = \mathcal{O}\left(\frac{1}{|s|^{1+\eta}}\right), \quad V'_{int}(s) = \mathcal{O}\left(\frac{1}{|s|^{2+\eta}}\right) \\ \text{and } V''_{int}(s) = \mathcal{O}\left(\frac{1}{|s|^{3+\eta}}\right) \text{ as } s \to \pm\infty. \end{aligned} \tag{1.4}$$

Remark 1.1. *Note that this forbids us to consider electrostatic interactions.*

We will also assume that the intra-chain potentials $\psi_1(s)$ and $\psi_\alpha(s)$ are smooth on $\mathbb{R} \setminus \{0\}$ and are bounded as well as their derivatives when s goes to $+\infty$.

Remark 1.2. *We could also easily study, e.g., convex potentials that blow up as s goes to $+\infty$. In this case, the minima $\varepsilon_{eq}(L)$ would remain bounded in some interval $[\varepsilon_{min}, \varepsilon_{max}]$. The estimates and convergence analysis that follows would then apply in this compact interval.*

We will further assume that $\psi_1(s)$ and $\psi_\alpha(s)$ blow up as $s \to 0$, i.e., for some $s_0 > 0$,

$$\forall s \in (-s_0, s_0), \qquad \psi_1(s), \psi_\alpha(\alpha s) \geq \frac{2}{|s|} \int_{\mathbb{R}} |V_{int}(t)| dt. \tag{1.5}$$

This technical assumption allows us to avoid situations where $\varepsilon_{eq}(L) \to -1$, i.e., the chains collapse, as shown by the following result.

Lemma 1.1. *If* (1.4) *and* (1.5) *are satisfied and L is large enough, there exists $\varepsilon_{min} > -1$ such that:*

$$\varepsilon_{min} \leq \varepsilon_{eq}(L), \tag{1.6}$$

where $\varepsilon_{eq}(L)$ is any minimizer of $\mathcal{E}(\varepsilon, L)$ defined by (1.2).

Proof. Since V_{int} is smooth and (1.4) is satisfied, the Riemann sum

$$(1+\varepsilon) \sum_{i \in \mathbb{Z}} |V_{int}\left(s - i(1+\varepsilon)\right)|$$

exists for all $s \in \mathbb{R}$ and converges uniformly to $\int_{\mathbb{R}} |V_{int}(t)| dt$ as $1+\varepsilon$ goes to zero. As a consequence, there exists $\varepsilon_0 > -1$ independent of L such that for $\varepsilon \in (-1, \varepsilon_0)$, we have for all $L \geq 1$:

$$\left| \sum_{i=0}^{[L]} V_{int}\left(s - i(1+\varepsilon)\right) \right| \leq \frac{2}{1+\varepsilon} \int_{\mathbb{R}} |V_{int}(t)| dt.$$

Replacing this estimate in (1.2) and using (1.5), we find that for $\varepsilon \in (-1, \min(\varepsilon_0, s_0 - 1))$ and L large enough,

$$\begin{aligned} \mathcal{E}(\varepsilon; L) &\geq \frac{2}{1+\varepsilon} \left(\frac{[L] + [\alpha^{-1}L]}{L} \int_{\mathbb{R}} |V_{int}(t)| dt - \frac{[\alpha^{-1}L] + 1}{L} \int_{\mathbb{R}} |V_{int}(t)| dt \right) \\ &\geq \frac{1}{1+\varepsilon} \int_{\mathbb{R}} |V_{int}(t)| dt. \end{aligned}$$

On the other hand, assumption (1.4) implies that $\mathcal{E}(0; L)$ is bounded from above for $L \geq 1$. Indeed, the series

$$s \mapsto \sum_{i=-\infty}^{\infty} |V_{int}\left(s - i\right)|$$

is absolutely convergent and defines a smooth 1-periodic function, in particular it is bounded. Substituting in (1.2), we find

$$\mathcal{E}(0; L) \leq \frac{1}{L} \left([L]\psi_1(1) + [\alpha^{-1}L]\psi_\alpha(\alpha) \right) + \frac{[\alpha^{-1}L] + 1}{L} \left\| \sum_{i=-\infty}^{\infty} |V_{int}\left(\cdot - i\right)| \right\|_\infty \leq C,$$

where $C > 0$ is some constant independent of L. Hence, there exists $\varepsilon_{min} > -1$ such that for $\varepsilon \in (-1, \varepsilon_{min})$, $\mathcal{E}(\varepsilon; L) > \mathcal{E}(0; L)$ for all L large enough. This proves (1.6). □

Now, due to Lemma 1.1, we can restrict our attention to those values of ε which belong to $[\varepsilon_{min}, \infty)$. Thanks to (1.4), the absolutely convergent series

$$V_{per} : (s, \varepsilon) \mapsto \sum_{i=-\infty}^{\infty} V_{int}\left((s-i)(1+\varepsilon)\right) \tag{1.7}$$

defines a smooth function $V_{per} \in C^2(\mathbb{R} \times [\varepsilon_{min}, \infty))$ which is 1-periodic in the s variable. We then consider the approximate periodized energy per unit length

$$\widetilde{\mathcal{E}}(\varepsilon; L) = \psi_1(1+\varepsilon) + \alpha^{-1}\psi_\alpha(\alpha + \varepsilon\alpha) + \alpha^{-1}\frac{1}{[\alpha^{-1}L]+1}\sum_{j=0}^{[\alpha^{-1}L]} V_{per}\left(\{\alpha j\}, \varepsilon\right), \tag{1.8}$$

where $\{\cdot\}$ denotes the fractional part of a real number. The following lemma shows that (1.8) defines a good approximation to the exact energy.

Lemma 1.2. *If* (1.4) *holds, there exists* $C > 0$ *independent of* L *and* ε *such that for* $\varepsilon > \varepsilon_{min}$ *and* $n \in \{0, 1, 2\}$*:*

$$\left|\frac{\partial^n \widetilde{\mathcal{E}}}{\partial \varepsilon^n}(\varepsilon; L) - \frac{\partial^n \mathcal{E}}{\partial \varepsilon^n}(\varepsilon; L)\right| \leq \begin{cases} C/L^\eta & \text{if } 0 < \eta < 1, \\ C \cdot \ln(L)/L & \text{if } \eta = 1, \\ C/L & \text{if } \eta > 1. \end{cases} \tag{1.9}$$

Proof. Thanks to (1.4), there exists $C > 0$ independent of L and ε such that the potential V_{per} and its first derivatives, $n \in \{0, 1, 2\}$, satisfy for $0 < s < L$:

$$\begin{aligned} &\left|\frac{\partial^n V_{per}}{\partial \varepsilon^n}(s, \varepsilon) - \sum_{i=0}^{[L]} (s-i)^n V_{int}^{(n)}\left((s-i)(1+\varepsilon)\right)\right| \\ &\qquad \leq \frac{C}{|1+\varepsilon|^{1+\eta}} \max\left(1, \frac{1}{|s|^\eta} + \frac{1}{|L-s|^\eta}\right). \end{aligned} \tag{1.10}$$

Taking the difference between (1.2) and (1.8) and using (1.10), we obtain immediately (1.9). □

As a consequence, the difference between the two energies $\widetilde{\mathcal{E}} - \mathcal{E}$ converges uniformly to zero in $C^2([\varepsilon_{min}, \infty))$ as $L \to \infty$. Therefore, it suffices to study the limit behavior of $\widetilde{\mathcal{E}}$ to determine the averaged elastic properties of the coupled system of chains in the limit $L \to \infty$. It is well-known [13] that the sequence $\{\alpha j\}_{j \in \mathbb{N}}$ is equidistributed in $[0, 1)$ for irrational α. This proves the following proposition.

Proposition 1.3 (Pointwise convergence of elastic energies)**.** *Let*

$$\mathcal{E}^\infty(\varepsilon) = \psi_1(1+\varepsilon) + \alpha^{-1}\psi_\alpha(\alpha + \varepsilon\alpha) + \alpha^{-1}\int_0^1 V_{per}\left(s, \varepsilon\right) ds. \tag{1.11}$$

Then, for all $\varepsilon \geq \varepsilon_{min}$*,* $n \in \{0, 1, 2\}$*,*

$$\lim_{L\to\infty} \frac{\partial^n \mathcal{E}}{\partial \varepsilon^n}(\varepsilon; L) = \lim_{L\to\infty} \frac{\partial^n \widetilde{\mathcal{E}}}{\partial \varepsilon^n}(\varepsilon; L) = \frac{\partial^n \mathcal{E}^\infty}{\partial \varepsilon^n}(\varepsilon). \tag{1.12}$$

The energy functional $\mathcal{E}^\infty(\varepsilon)$ can thus be considered as the Cauchy-Born elastic energy density of the coupled system of chains, relating the strain ε to the potential energy of the system.

2 Convergence analysis

The convergence result obtained in Proposition 1.3 is not enough to ensure either the uniform convergence of the energy functionals $\mathcal{E}$ and $\widetilde{\mathcal{E}}$, nor the convergence of their minimizers (Γ-convergence). To prove these statements, and to obtain more precise error estimates useful e.g. for the numerical computation of the quantities of interest at the macroscopic level, we need some results from discrepancy theory which we recall in the following.

2.1 A primer on discrepancy and Birkhoff sums

Let $\omega = (x_n)_{n\geq 1}$ be a given sequence of real numbers in I. For a positive integer N and a subset E of I, let the counting function $A(E; N; \omega)$ be defined as the number of terms x_n for $1 \leq n \leq N$ for which $\{x_n\} \in E$. Where no confusion is possible, we write $A(E; N)$ instead of $A(E; N; \omega)$.

Definition 2.1 (Discrepancy)**.** *Let $x_1, \ldots, x_N$ be a finite sequence of real numbers in I. The number*

$$D_N = D_N(x_1, \ldots, x_N) = \sup_{0\leq\alpha<\beta\leq 1} \left| \frac{A([\alpha, \beta); N)}{N} - (\beta - \alpha) \right| \tag{2.1}$$

is called the discrepancy of the given sequence. For an infinite sequence ω of real numbers, or for a finite sequence containing at least N terms, the discrepancy $D_N(\omega)$ is meant to be the discrepancy of the initial segment formed by the first N terms of ω.

An alternate definition is given by

Definition 2.2. *For a finite sequence of real numbers $x_1, \ldots, x_N$ in I, we define*

$$D_N^* = D_N^*(x_1, \ldots, x_N) = \sup_{0<\alpha\leq 1} \left| \frac{A([0, \alpha); N)}{N} - \alpha \right|. \tag{2.2}$$

This definition is extended as the previous one to infinite sequences. The discrepancies D_N and D_N^* are related by the following inequality, see [13]:

$$D_N^* \leq D_N \leq 2D_N^*. \tag{2.3}$$

Theorem 2.3 (Koksma's inequality, [13])**.** *Let ϕ be a function on I of bounded variation $\|\phi\|_{TV}$, and suppose we are given N points $x_1, \ldots, x_N$ in I with discrepancy D_N^*. Then,*

$$\left| \frac{1}{N} \sum_{i=1}^{N} \phi(x_i) - \int_0^1 \phi(x)dx \right| \leq \|\phi\|_{TV} D_N^*. \tag{2.4}$$

In general, the discrepancy of the sequence generated by iterating an irrational rotation, $x_i = \{i\alpha\} \in \mathbb{T}$ with $\alpha \in \mathbb{R} \setminus \mathbb{Q}$, goes to zero as N goes to infinity thanks to the ergodic theorem. However this rate of convergence could be arbitrarily slow and depends in general strongly on the number theoretical properties of α. A more precise convergence rate can be deduced in an average sense from the following result owing to Kesten [11]:

Theorem 2.4. *Let $D_N(\alpha)$ be the discrepancy of the sequence $(\{i\alpha\})_{0\leq i<N} \subset \mathbb{T}$ for $\alpha \in [0,1]$, $N \in \mathbb{N}$. Then:*

$$\frac{N \cdot D_N(\alpha)}{\log N \cdot \log\log N} \to \frac{2}{\pi^2} \text{ in measure on } [0,1] \text{ as } N \to \infty. \tag{2.5}$$

To obtain pointwise estimates, e.g. to bound the error in numerical computations, it is necessary to choose carefully N, the number of atoms in our model. For irrational α, we say that $\frac{p}{q} \in \mathbb{Q}$ is a rational approximation of α if $p \in \mathbb{Z}$, $q \in \mathbb{N}$, $p \wedge q = 1$, and

$$\left|\alpha - \frac{p}{q}\right| < \frac{1}{q^2}. \tag{2.6}$$

For irrational α, an infinite number of such rational approximations exist and they can be obtained as the convergents from its continued fraction expansion, see [10].

Remark 2.1. *Note that the following result also allows to tackle the general case of homeomorphisms of the circle which are not necessarily rigid rotations, a situation which appears in relaxed configurations as will be studied in a second paper [5].*

Theorem 2.5 (Denjoy-Koksma inequality [10])**.** *Let f be an orientation-preserving homeomorphism of $\mathbb{T}$ with rotation number $\rho(f) = \alpha \in \mathbb{R}\setminus\mathbb{Q}$, and p/q a rational approximation of α. Let $\phi : \mathbb{T} \mapsto \mathbb{R}$ be a function with bounded variation $\|\phi\|_{TV}$, not necessarily continuous, and let μ be a probability measure on $\mathbb{T}$ invariant by f (i.e. $f_*\mu = \mu$). Then, for all $x \in \mathbb{T}$, we have:*

$$\left|\sum_{i=0}^{q-1} \phi \circ f^i(x) - q\int_0^1 \phi(x)d\mu\right| \leq \|\phi\|_{TV}. \tag{2.7}$$

In fact, an improved result is obtained for continuous functions, if we restrict ourselves to the situation of a rigid rotation:

Proposition 2.6 (Improved Denjoy-Koksma inequality [10])**.** *Let R_α be an irrational rotation of $\mathbb{T}$, and $\frac{p}{q}$ a rational approximation of α. Let $\phi : \mathbb{T} \mapsto \mathbb{R}$ be an absolutely continuous function, and w its continuity modulus. Then, for all $x \in \mathbb{T}$, we have:*

$$\left|\frac{1}{q}\sum_{i=0}^{q-1} \phi \circ R_\alpha^i(x) - \int_0^1 \phi(x)d\mu\right| \leq w\left(\frac{1}{q}\right). \tag{2.8}$$

More precisely, we have the following result:

Proposition 2.7 ([10]). *Let R_α be an irrational rotation of $\mathbb{T}$, and $\frac{p_n}{q_n}$ the sequence of rational approximations of α obtained from its continued fraction expansion. Then if $\phi : \mathbb{T} \mapsto \mathbb{R}$ is an absolutely continuous function, we have*

$$\left\| \sum_{i=0}^{q_n-1} \phi \circ R_\alpha^i - q_n \int_0^1 \phi(x)dx \right\|_{C^0} \to 0 \qquad \text{as } n \to \infty. \tag{2.9}$$

The convergence rate can be improved if we assume some more regularity:

Proposition 2.8. *Let R_α be an irrational rotation of $\mathbb{T}$ and $\frac{p}{q}$ a rational approximation of α. Then if $\phi : \mathbb{T} \mapsto \mathbb{R}$ is differentiable and its derivative is a function with bounded variation, we have:*

$$\left\| \sum_{i=0}^{q-1} \phi \circ R_\alpha^i - q \int_0^1 \phi(x)dx \right\|_{C^0} \leq \frac{2}{q} \left\| \phi' \right\|_{TV}. \tag{2.10}$$

Proof. It is enough to consider $\phi : \mathbb{T} \mapsto \mathbb{R}$ with zero average, such that ϕ' has bounded variation $\|\phi'\|_{\mathrm{TV}}$. Let us denote

$$\phi_q = \sum_{i=0}^{q-1} \phi \circ R_\alpha^i.$$

Since ϕ' has mean value zero,

$$\|\phi'\|_\infty = \max_{x \in \mathbb{T}} \left| \int_0^1 \phi(x) - \phi(y)dy \right| \leq \left\| \phi' \right\|_{\mathrm{TV}},$$

and by the Denjoy-Koksma inequality,

$$\left\| \phi_q' \right\|_\infty = \left\| \sum_{i=0}^{q-1} \phi' \circ R_\alpha^i \right\|_\infty \leq \left\| \phi' \right\|_{\mathrm{TV}}.$$

We notice that for any $x \in \mathbb{T}$,

$$|\phi_q(x+\alpha) - \phi_q(x)| = |\phi(x+q\alpha) - \phi(x)| \leq \left\| \phi' \right\|_\infty |q\alpha - p| \leq \frac{\|\phi'\|_{\mathrm{TV}}}{q^2}. \tag{2.11}$$

By the mean value property, there exists $x_0 \in \mathbb{T}$ such that $\phi_q(x_0) = 0$. By (2.11), we deduce that

$$|\phi_q(x_0 + j\alpha)| \leq \frac{j}{q^2} \left\| \phi' \right\|_{\mathrm{TV}} \leq \frac{\|\phi'\|_{\mathrm{TV}}}{q}, \qquad \forall 1 \leq j \leq q. \tag{2.12}$$

Now for any interval $I_i = [\frac{i}{q}, \frac{i+1}{q}]$, $i = 0 \dots q-1$, there exists a unique $j \in 1 \dots q$ such that $j\alpha \in I_i$, see e.g. [10] V.8.1. Then for any $x \in \mathbb{T}$, there exists $j \in 1 \dots q$ such that $x - x_0 \in I_i$ and $j\alpha \in I_i$, and thus

$$|\phi_q(x) - \phi_q(x_0 + j\alpha)| \leq \left\| \phi' \right\|_\infty |x - (x_0 + j\alpha)| \leq \frac{\|\phi'\|_{\mathrm{TV}}}{q}. \tag{2.13}$$

Combining (2.12) and(2.13), we obtain the desired result:

$$\forall x \in \mathbb{T}, \qquad |\phi_q(x)| < \frac{2}{q} \left\|\phi'\right\|_{\mathrm{TV}}.$$

□

We present in Fig. 2 a graphical representation of the behavior of the Birkhoff sums measured by the deviation

$$\delta_q(\phi) = \left| \sum_{i=0}^{q-1} \phi \circ R_\alpha^i(x) - q \int_0^1 \phi(x) d\mu \right|$$

when α is chosen as the golden mean. These results indicate that the Denjoy-Koksma inequalities (2.7) and (2.8) give a sharp estimate of the effective convergence rate.

- For a discontinuous function, δ_q is bounded from above but does not decrease as in Fig. 2(a);
- For an absolutely continuous function, δ_q converges to zero. If the derivative of the function has bounded variation as in Fig. 2(c), then the rate of convergence is linear as $1/q$, otherwise it is only sub-linear as in Fig. 2(b).

In all cases, we observe that the behavior for general N appears chaotic with oscillations spanning orders of magnitude. To achieve accurate and efficient numerical computations, it thus appears necessary to use criterion (2.6) to choose an appropriate finite number of atoms N in our experiments, at least for this elementary problem of computing averaged quantities. Equivalently, good choices for q are obtained from the continued fraction expansion of α.

We will further show in [5] that this choice also allows for accurate predictions in a relaxed state, involving optimization of the positions of the atoms.

2.2 Error estimates

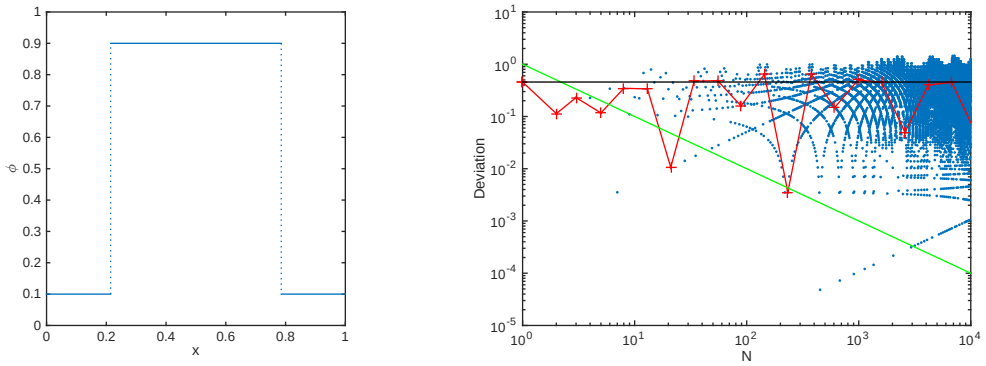
Using these results, it is straightforward to obtain formulas for the averaged properties of the double chain model from Section 1 and study the rate of convergence.

First, thanks to Koskma's inequality (2.4) and the fact that the discrepancy of the sequence $\{\alpha j\}_{0 \le j \le [\alpha^{-1} L]}$ goes to zero as $L \to \infty$, we obtain immediately the uniform convergence of the elastic energies defined in (1.2), (1.8) and (1.11):

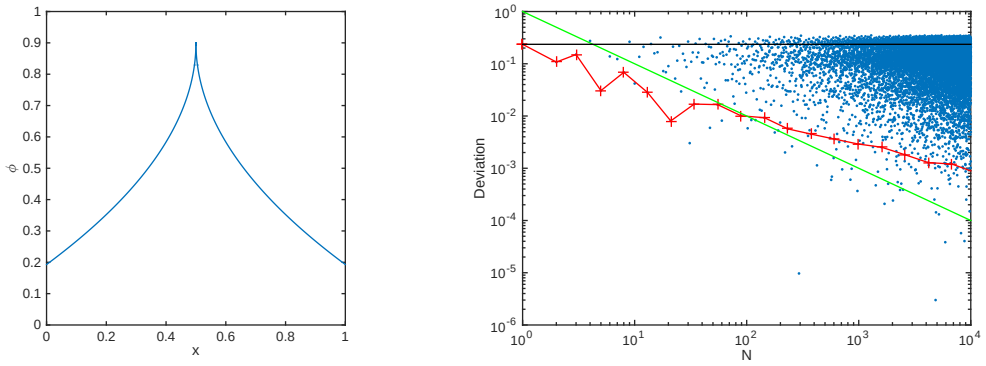
Theorem 2.9. *Under assumptions* (1.4) *and* (1.5)*, we have:*

$$\begin{cases} \mathcal{E}(\varepsilon; L) \to \mathcal{E}^\infty(\varepsilon) \\ \widetilde{\mathcal{E}}(\varepsilon; L) \to \mathcal{E}^\infty(\varepsilon) \end{cases} \text{ as } L \to \infty, \text{ uniformly in } C^1([\varepsilon_{min}, \infty)). \tag{2.14}$$

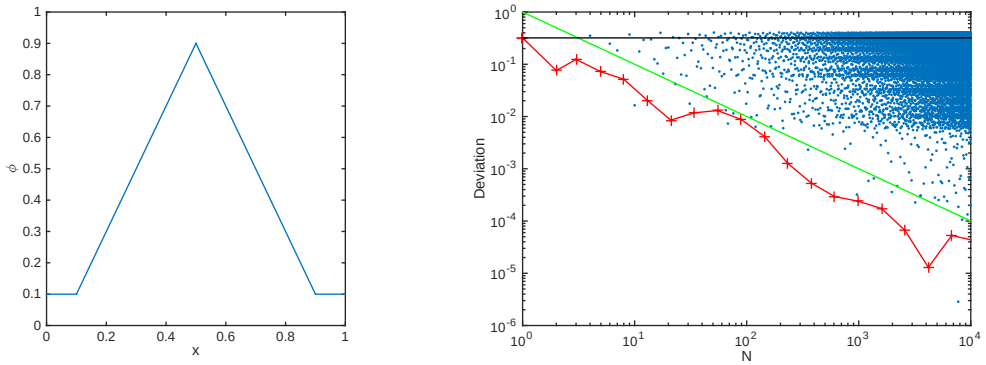
Remark 2.2. *Note that the rate of convergence in* (2.14) *could be arbitrarily slow.*



(a) Discontinuous ϕ



(b) Absolutely continuous, non-Lipschitz ϕ



(c) Lipschitz ϕ

Figure 2: Convergence behavior of the Birkhoff sums for various choices of ϕ and α chosen as the golden mean. In each case, we plot on the left the function ϕ on $[0, 1]$. On the right is the log-plot for the deviation of the sum from the asymptote (left-hand term in (2.7). The values obtained for general N is in blue, in red we plot the values for $N = q$ satisfying (2.6), in green the optimal convergence rate N^{-1}, in black a constant.

Proof. To show (2.14), we need only show that $\|V_{per}(\cdot,\varepsilon)\|_{\mathrm{TV}}$ and $\left\|\frac{\partial}{\partial\varepsilon}V_{per}(\cdot,\varepsilon)\right\|_{\mathrm{TV}}$ in $[0,1]$ are bounded for $\varepsilon \geq \varepsilon_{min}$. Then Koskma's inequality (2.4) is sufficient to finish the proof.

Now, from (1.7) we deduce:

$$\begin{aligned}\|V_{per}(\cdot,\varepsilon)\|_{\mathrm{TV}} &= \int_0^1 \left|\frac{\partial}{\partial s}V_{per}(s,\varepsilon)\right| ds \\ &\leq \int_0^1 \sum_{i\in\mathbb{Z}} \left|(1+\varepsilon)V'_{int}((s-i)(1+\varepsilon))\right| ds.\end{aligned}$$

By a change of variables, this leads to

$$\|V_{per}(\cdot,\varepsilon)\|_{\mathrm{TV}} \leq \int_{\mathbb{R}} \left|V'_{int}(s)\right| ds. \tag{2.15}$$

Note that $\|V_{int}\|_{\mathrm{TV}} = \int_{\mathbb{R}} |V'_{int}(s)|\, ds$ is finite because of the bounds (1.4). Hence $\|V_{per}(\cdot,\varepsilon)\|_{\mathrm{TV}}$ is uniformly bounded. Similarly,

$$\begin{aligned}\left\|\frac{\partial}{\partial\varepsilon}V_{per}(\cdot,\varepsilon)\right\|_{\mathrm{TV}} &= \int_0^1 \left|\frac{\partial^2}{\partial s\partial\varepsilon}V_{per}(s,\varepsilon)\right| ds \\ &\leq \int_0^1 \sum_{i\in\mathbb{Z}} \left|(1+\varepsilon)(s-i)V''_{int}((s-i)(1+\varepsilon))\right| + \sum_{i\in\mathbb{Z}} \left|V'_{int}((s-i)(1+\varepsilon))\right| ds \\ &\leq \frac{1}{1+\varepsilon}\left(\int_{\mathbb{R}} \left|sV''_{int}(s)\right| ds + \int_{\mathbb{R}} \left|V'_{int}(s)\right| ds\right).\end{aligned}$$

Because of the bounds (1.4), $\int_{\mathbb{R}} |sV''_{int}(s)|\, ds$ is finite, and this concludes the proof. □

Corollary 2.10. *The sequence of energy functionals $\varepsilon \mapsto \widetilde{\mathcal{E}}(\varepsilon;L)$ Γ-converges to $\mathcal{E}^\infty$ as $L\to\infty$. In particular, any converging sequence of minimizers $\widetilde{\varepsilon_{eq}}(L)$ of $\mathcal{E}(\cdot;L)$ converges to a minimizer ε_{eq}^∞ of $\mathcal{E}^\infty$, and the associated elastic constants converge:*

$$\widetilde{\mathcal{K}}(\widetilde{\varepsilon_{eq}};L) = \frac{\partial^2\widetilde{\mathcal{E}}}{\partial\varepsilon^2}(\widetilde{\varepsilon_{eq}}(L);L) \to \mathcal{K}^\infty(\varepsilon_{eq}^\infty) = \frac{\partial^2\mathcal{E}^\infty}{\partial\varepsilon^2}(\varepsilon_{eq}^\infty). \tag{2.16}$$

Remark 2.3. *The same results hold also for $\mathcal{E}(\varepsilon;L)$, an associated converging sequence of minimizers $\varepsilon_{eq}(L)$ and the associated elastic constants $\mathcal{K}(\varepsilon_{eq};L)$ thanks to* (2.14).

Remark 2.4. *In particular, if the Cauchy-Born energy density $\mathcal{E}_{eq}^\infty$ has a unique minimum at $\varepsilon_{eq}^\infty \in (\varepsilon_{min},\infty)$ and the minimizers of $\varepsilon\mapsto\mathcal{E}(\cdot;L)$ belong to a bounded set $(\varepsilon_{min},\varepsilon_{max})$ independently of L, then any sequence of minimizers converges to ε_{eq}^∞ and the elastic constants associated with these minimizers also converge to $\mathcal{K}^\infty(\varepsilon_{eq}^\infty)$.*

Proof. The uniform convergence of the sequence of functionals $\varepsilon\mapsto\mathcal{E}(\varepsilon;L)$ to the continuous functional $\mathcal{E}^\infty$ implies its Γ-convergence, see e.g. [6]. The Fundamental Theorem of Γ-convergence then implies that the limit of a converging sequence of minimizers $\varepsilon_{eq}(L)$ of $\mathcal{E}(\cdot;L)$ is a minimizer ε_{eq}^∞ of $\mathcal{E}^\infty$.

Since $\frac{\partial^2 \widetilde{\mathcal{E}}}{\partial \varepsilon^2}$ does not necessarily converge uniformly, we cannot deduce (2.16) directly. By (1.12), we have

$$\lim_{L\to\infty} \frac{\partial^2 \widetilde{\mathcal{E}}}{\partial \varepsilon^2}(\varepsilon_{eq}^{\infty}; L) = \frac{\partial^2 \mathcal{E}^{\infty}}{\partial \varepsilon^2}(\varepsilon_{eq}^{\infty}). \tag{2.17}$$

However, $\frac{\partial^2}{\partial \varepsilon^2} V_{per}$ is uniformly continuous in a neighborhood of $[0,1] \times \{\varepsilon_{eq}^{\infty}\}$, so for all $\delta > 0$ there exists $\gamma > 0$ such that if $|\varepsilon_{eq}(L) - \varepsilon_{eq}^{\infty}| < \gamma$,

$$\frac{1}{L} \left| \sum_{j=0}^{[\alpha^{-1}L]} \frac{\partial^2}{\partial \varepsilon^2} V_{per}\left(\{\alpha j\}, \varepsilon_{eq}^{\infty}\right) - \sum_{j=0}^{[\alpha^{-1}L]} \frac{\partial^2}{\partial \varepsilon^2} V_{per}\left(\{\alpha j\}, \varepsilon_{eq}(L)\right) \right| < \delta.$$

Hence, considering (1.8), (2.17) and since $\varepsilon_{eq}(L) \to \varepsilon_{eq}^{\infty}$, there exists $L_\delta > 0$ such that

$$L > L_\delta \implies \left| \frac{\partial^2 \widetilde{\mathcal{E}}}{\partial \varepsilon^2}(\varepsilon_{eq}(L); L) - \frac{\partial^2 \mathcal{E}^{\infty}}{\partial \varepsilon^2}(\varepsilon_{eq}^{\infty}) \right| < 2\delta.$$

This proves (2.16). □

Note that we have obtained in this one-dimensional toy model a closed-form expression (1.11) for the 'macroscopic' (limit) energy $\mathcal{E}_{eq}^{\infty}$. In more complex systems, for example in the relaxed configurations we will study in a forthcoming paper [5], such explicit formulae do not exist. It is then necessary to employ numerical simulations to determine the macroscopic energy density. When dealing with commensurate systems, this typically leads to solving so-called corrector problems which are set on a unit cell of the commensurate lattice, using periodic boundary conditions. However for incommensurate systems, such unit cells do not exist and the corrector problem, if it is well-posed, is set on the whole space.

Remark 2.5. *The same situation arises for example in homogenization of PDEs with periodic vs. stationary random coefficients.*

It is therefore instructive to study the convergence rate of the energy density $\widetilde{\mathcal{E}}(\varepsilon; L)$ as $L \to \infty$. By comparing the respective definitions (1.8) and (1.11), we see that this amounts to studying the convergence rate of the limit

$$\frac{1}{N} \sum_{j=0}^{N-1} V_{per}\left(\{\alpha j\}, \varepsilon\right) \to \int_0^1 V_{per}\left(s, \varepsilon\right) ds \quad \text{as } N \to \infty, \tag{2.18}$$

where $N = [\alpha^{-1} L]$ is the number of atoms in the $\mathcal{C}^{\alpha}$ layer.

Remark 2.6. *Note that by studying $\widetilde{\mathcal{E}}(\varepsilon; L)$ instead of the exact energy $\mathcal{E}(\varepsilon; L)$, we remove boundary effects limiting the convergence rate to $\frac{1}{L}$ at best, see Lemma 1.2.*

Now the left hand term in (2.18) is a Birkhoff sum. The classical results summarized in Section 2.1 enable us to make the following statements:

- From Theorems 2.3 and 2.4, we deduce that the measure of the set of $\alpha \in (0,1)$ such that, for any $\varepsilon > \varepsilon_{min}$,

$$\left| \frac{1}{N} \sum_{j=0}^{N-1} V_{per}\left(\{\alpha j\}, \varepsilon\right) - \int_0^1 V_{per}\left(s, \varepsilon\right) ds \right| > \frac{2 \left\| V_{per}(\cdot, \varepsilon)) \right\|_{\mathrm{TV}}}{\pi^2} \frac{\log N \cdot \log\log N}{N}$$

goes to zero as $N \to \infty$. Hence, 'on average', we expect to observe a convergence rate close to $\frac{1}{N}$. Still, for a vanishing set of α values the left-hand side could converge arbitrarily slow.

- If N is chosen as the denominator q of a convergent of α, i.e.,

$$p, q \in \mathbb{N} \text{ such that } \left| \alpha - \frac{p}{q} \right| < \frac{1}{q^2},$$

then by Theorem 2.6, for all $\varepsilon > \varepsilon_{min}$,

$$\left| \frac{1}{q} \sum_{j=0}^{q-1} V_{per}\left(\{\alpha j\}, \varepsilon\right) - \int_0^1 V_{per}\left(s, \varepsilon\right) ds \right| < \frac{2}{q^2} \left\| \frac{\partial V_{per}}{\partial s}(\cdot, \varepsilon) \right\|_{\mathrm{TV}}.$$

Note that similar convergence rates can be obtained for $\frac{\partial}{\partial \varepsilon} V_{per}$ if its derivative against s has bounded variation.

We have thus obtained rigorous convergence rates for both $\widetilde{\mathcal{E}}(\varepsilon; L)$ and $\frac{\partial}{\partial \varepsilon}\widetilde{\mathcal{E}}(\varepsilon; L)$. Moreover, assuming that

$$\frac{\partial^2 \mathcal{E}^\infty}{\partial \varepsilon^2}(\varepsilon_{eq}^\infty) > 0,$$

it is readily seen that whenever a sequence of minimizers $\{\varepsilon_{eq}\}_{L>0}$ converges to ε_{eq}^∞, the convergence rate is the same as observed for the energies. Therefore, it is also the case e.g. for the linearized Cauchy-Born elastic constant around the ground state.

To conclude the analysis of convergence for this toy model, let us point out that the direct computation by simply increasing the number of atoms N achieves a nearly linear rate of convergence *on average*, but a careful choice of the sample size yields a quadratic convergence rate *every time*. This quite unusual result is a consequence of the incommensurability of the system.

3 Approximation by periodic configurations

Finally, let us construct and analyze a second approximate model based on approximating the incommensurate system by a sequence of periodic configurations. This study is interesting as an elementary example of this very common approach to the computational modeling of incommensurate composites, see e.g. [19]. While everything is explicit in this one-dimensional example, the analysis will be extended in future papers to study the elastic relaxation of incommensurate double chain models.

Let us take a sequence of rational approximations of α, i.e., pairs of mutually prime integers p_n, q_n such that:

$$\frac{p_n}{q_n} \xrightarrow[n\to\infty]{} \alpha.$$

We now study the convergence of the energy of configurations where the ratio of the atom spacing in the second chain compared to the first is p_n/q_n instead of α. Interactions between atoms are given by the same potentials ψ_1, ψ_α and V_{int} as before. These new configurations can now be made periodic, such that the period at rest is p_n. Over one period, the first and second chain contain respectively p_n and q_n atoms. Furthermore, the energy per unit length can be written similarly to the previous periodized energy (1.8):

$$\widetilde{\mathcal{E}}_n(\varepsilon) = \psi_1(1+\varepsilon) + \frac{q_n}{p_n}\cdot\psi_\alpha\left((1+\varepsilon)\frac{p_n}{q_n}\right) + \frac{q_n}{p_n}\frac{1}{q_n}\sum_{j=0}^{q_n-1} V_{per}\left(\left\{j\frac{p_n}{q_n}\right\},\varepsilon\right). \tag{3.1}$$

Note that this energy is obtained by averaging over one period. This effectively amounts to taking the limit $L\to\infty$ in this periodic setting. We can further simplify (3.1) by observing that since p_n and q_n are mutually prime, we have equality between discrete sets, up to some reordering:

$$\left(\left\{j\frac{p_n}{q_n}\right\}\right)_{0\le j<q_n} = \left(\frac{j}{q_n}\right)_{0\le j<q_n}.$$

Therefore,

$$\widetilde{\mathcal{E}}_n(\varepsilon) = \psi_1(1+\varepsilon) + \frac{q_n}{p_n}\cdot\psi_\alpha\left((1+\varepsilon)\frac{p_n}{q_n}\right) + \frac{1}{p_n}\sum_{j=0}^{q_n-1} V_{per}\left(\frac{j}{q_n},\varepsilon\right). \tag{3.2}$$

Remark 3.1. *By modifying the lattice constant in the second chain, we have obtained a formula which involves only a finite number of atoms. For more complex systems, this allows one to compute easily relaxed configurations or electronic properties. The price for this simplification is that we have introduced some additional, artificial strain in the system.*

Proposition 3.1. *Under assumptions* (1.4) *and* (1.5)*, we have the error estimate:*

$$\left|\widetilde{\mathcal{E}}_n(\varepsilon) - \widetilde{\mathcal{E}}^\infty(\varepsilon)\right| \le C(1+\varepsilon)^2\left(\frac{1}{q_n^2} + \left|\frac{p_n}{q_n}-\alpha\right|\right). \tag{3.3}$$

Remark 3.2. *Note that, unlike the discrepancy-based estimates constructed for the exact model in section 2.2, the rate of convergence cannot be arbitrarily slow as q_n goes to infinity. Note that the optimal convergence rate $\mathcal{O}(1/q_n^2)$ is again recovered for the convergents of α.*

Proof. The last term in the definition (3.2) is simply a Riemann sum. Recall that $\widetilde{\mathcal{E}}^{\infty}(\varepsilon)$ is given by (1.11). Let us then bound by considering separately each term in the error:

$$\left|\widetilde{\mathcal{E}}_n(\varepsilon) - \widetilde{\mathcal{E}}^{\infty}(\varepsilon)\right| \leq \left|\frac{q_n}{p_n} \cdot \psi_\alpha\left((1+\varepsilon)\frac{p_n}{q_n}\right) - \frac{1}{\alpha} \cdot \psi_\alpha\left((1+\varepsilon)\alpha\right)\right| + \left|\frac{1}{p_n}\sum_{j=0}^{q_n-1} V_{per}\left(\frac{j}{q_n}, \varepsilon\right) - \frac{1}{\alpha}\int_0^1 V_{per}(s,\varepsilon)ds\right|.$$

First, we have for some constant $C > 0$:

$$\left|\frac{q_n}{p_n} \cdot \psi_\alpha\left((1+\varepsilon)\frac{p_n}{q_n}\right) - \frac{1}{\alpha} \cdot \psi_\alpha\left((1+\varepsilon)\alpha\right)\right| \leq C(1+\varepsilon)\left|\frac{q_n}{p_n} - \alpha\right|, \tag{3.4}$$

where we use the fact that ψ_α and ψ'_α are bounded on the interval $[\varepsilon_{min}, \infty)$. Next, to evaluate the Riemann sum, we make use of the estimate

$$\left|\frac{\partial^2 V_{per}}{\partial s^2}(s,\varepsilon)\right| \leq C(1+\varepsilon)^2,$$

where $C > 0$ is a constant independent of ε and s. This bound can be derived directly from the definition (1.7) of the periodic potential under the assumptions (1.4). Then, using the standard error estimate, we obtain:

$$\left|\frac{1}{q_n}\sum_{j=0}^{q_n-1} V_{per}\left(\frac{j}{q_n}, \varepsilon\right) - \int_0^1 V_{per}(s,\varepsilon)ds\right| \leq C\frac{(1+\varepsilon)^2}{q_n^2}. \tag{3.5}$$

To conclude, we observe that the sequence p_n/q_n is bounded, and also

$$\left|\int_0^1 V_{per}(s,\varepsilon)ds\right| \leq \int_0^1 \sum_{i\in\mathbb{Z}} |V_{int}((s-i)(1+\varepsilon))| \leq \frac{1}{1+\varepsilon}\int_{\mathbb{R}} |V_{int}(S)|\, dS, \tag{3.6}$$

where we use the change of variables $S \equiv (1+\varepsilon)(s-i)$. Bringing together estimates (3.4), (3.5) and (3.6) we find the desired result (3.3). □

4 Conclusion

In this work, we have studied a Cauchy-Born-type energy density for a coupled system of one-dimensional incommensurate coupled chains. We have shown that it is given by a closed-form formula, Eq. (1.11), in the thermodynamic limit of infinite system size. This allows to study rigorous estimates of the convergence rate, as a function of either the total length of the system or for periodic approximants to be used in numerical computations.

This study provides a rigorous theoretical foundation for the modeling of incommensurate heterostructures composed of monolayers such as graphene, boron nitride or molybdenum disulfide. Work is ongoing on the further development and analysis of more realistic models, including effects such as out-of-plane relaxation of the lattices forming ripples [5].

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