PAPER

Duality relations for the classical ground states of soft-matter systems

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Bounded interactions are particularly important in soft-matter systems, such as colloids, microemulsions, and polymers. In this paper, we extend the results of a recent letter [S. Torquato and F. H. Stillinger, *Phys. Rev. Lett.*, 2008, **100**, 020602] on duality relations for ground states of pair interactions to include three-body and higher-order functions. Our novel and general relations link the energy of configurations associated with a real-space potential to the corresponding energy of the dual (Fourier-transformed) potential and can be applied to ordered and disordered classical ground states. We use the duality relations to demonstrate how information about the classical ground states of short-ranged potentials can be used to draw new conclusions about the ground states of long-ranged potentials and *vice versa*. The duality relations also lead to bounds on the $T = 0$ system energies in density intervals of phase coexistence. Additionally, we identify classes of "self-similar" potentials, for which one can rigorously relate low- and high-density ground-state energies. We analyze the ground state configurations and thermodynamic properties of a one-dimensional system expected to exhibit an infinite number of structural phase transitions and comment on the known ground states of purely repulsive monotonic potentials in the context of our duality relations.

1. Introduction

The zero-temperature ground states of classical interacting many-particle systems are minimal-energy configurations, typically corresponding to crystalline Bravais lattices. Although numerical simulations and experiments that slowly freeze liquids can probe ground-state structures, developing a theoretical framework for identifying these configurations remains an open problem in condensed-matter physics and materials science.[1,2] The relatively simple topology of the line has allowed much progress to be made toward identifying ground states of certain interactions for lattice models;[2] unfortunately, solutions in d-dimensional Euclidean space $\mathbb{R}^d$ for $d \geq 2$ are considerably more challenging. For example, the ground state(s) of the well-known Lennard-Jones potential in $\mathbb{R}^2$ or $\mathbb{R}^3$ are not known rigorously.[3] New theoretical tools are therefore necessary in order to understand these more difficult problems. Recent research toward this end has used a "collective coordinate" approach to explore the ground states in two and three dimensions for a certain class of interactions.[4,5] Surprisingly, nontrivial *disordered* classical ground states without any long-range order have been identified in these dimensions[4,6] along with the expected periodic ones.

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In a recent Letter, we derived duality relations for a certain class of soft pair potentials that can be applied to classical ground states whether they are disordered or not.[7] Soft, bounded interactions are particularly important in soft-matter systems, such as colloids, microemulsions, and polymers.[8–11] Additionally, since these potentials lack non-integrable singularities, they are easier to treat theoretically. The duality relations discussed in ref. 7 link the energy of configurations associated with a real-space pair potential $v(r)$ to the energy associated with the dual (Fourier-transformed) potential. Such relations are useful because they enable one to use information about the ground states of soft short-ranged potentials to draw conclusions about the ground states of long-ranged potentials and *vice versa*. In particular, these relations aid in numerical simulations of ground-state structures as described in Section III. Furthermore, the duality relations provide rigorous bounds on the ground state energies in density intervals of phase coexistence.

Here we provide novel duality relations that apply for general n-particle interactions. Our generalized relations allow one to examine the ground states of soft-matter systems that are not sufficiently described by pair potentials. Indeed, the inclusion of higher-order interactions can have a significant effect on the observed ground-state structures as has been shown in recent work on the covering and quantizer problems from discrete mathematics, information theory, and number theory.[12] Additionally, we introduce a general class of potential functions that are "self-similar" under Fourier transform; for any of the potentials in this class, one can rigorously relate the low- and high-density ground state energies. Our results are applied to

a one-dimensional system that was argued to possess an infinite number of structural phase transitions from Bravais to non-Bravais lattices at $T = 0$ as the density is changed.[7] We analytically show that there is a high degeneracy of ground-state configurations, and the thermodynamic properties of this system, including the entropy and thermal expansion coefficient, likely vary nonanalytically at integer-valued densities. Furthermore, we comment on the known ground states of purely repulsive monotonic potentials in the context of our duality relations, including cases where non-Bravais lattices and their "formal duals" (defined below) appear as ground-states.

Section II provides the requisite background on point processes, Bravais and non-Bravais lattices, and (formally) dual periodic structures. Section III briefly reviews the key results of ref. 7 by discussing duality relations for pair interactions and then extends the methodology of that paper to incorporate three-body and higher-order potentials. Section IV presents applications of our relations for functions with compact support, nonnegative potentials, and completely monotonic potentials. Specific attention is given to the aforementioned one-dimensional many-particle system and its ground state structural and thermodynamic properties. Section V describes a class of interactions that are self-dual under Fourier transform, including the special case of pair potentials that are eigenfunctions of the Fourier transform. Discussion and concluding remarks are in Section VI.

II.   Definitions and preliminaries

A point process in $\mathbb{R}^d$ is a distribution of an infinite number of points at number density ρ (number of points per unit volume) with configuration $\mathbf{r}_1, \mathbf{r}_2, \ldots$; see ref. 12 for a precise mathematical definition. It is characterized by a countably infinite set of n-particle generic probability density functions $\rho_n(\mathbf{r}_1, \ldots, \mathbf{r}_n)$, which are proportional to the probability densities of finding collections of n particles in volume elements near the positions $\mathbf{r}_1, \ldots, \mathbf{r}_n$. For a general point process, it is convenient to introduce the n-particle correlation functions g_n, which are defined by

$$g_n(\mathbf{r}_1, \ldots, \mathbf{r}_n) = \frac{\rho_n(\mathbf{r}_1, \ldots, \mathbf{r}_n)}{\rho^n}. \tag{1}$$

Since $\rho_n = \rho^n$ for a completely uncorrelated point process, it follows that deviations of g_n from unity provide a measure of the correlations between points in a point process. Of particular interest is the pair correlation function, which for a translationally invariant point process of density ρ can be written as

$$g_2(\mathbf{r}) = \frac{\rho_2(\mathbf{r})}{\rho^2} \qquad (\mathbf{r} = \mathbf{r}_2 - \mathbf{r}_1). \tag{2}$$

Closely related to the pair correlation function is the *total correlation function*, denoted by h; it is derived from g_2 *via* the equation

$$h(\mathbf{r}) = g_2(\mathbf{r}) - 1. \tag{3}$$

Since $g_2(r) \to 1$ as $r \to +\infty$ ($r = |\mathbf{r}|$) for translationally invariant systems without long-range order, it follows that $h(r) \to 0$ in this limit, meaning that h is generally an L^2 function, and its Fourier transform is well-defined.

It is common in statistical mechanics when passing to reciprocal space to consider the associated *structure factor* S, which for a translationally invariant system is defined by

$$S(k) = 1 + \rho \tilde{h}(k), \tag{4}$$

where $\tilde{h}$ is the Fourier transform of the total correlation function, ρ is the number density, and $k = |\mathbf{k}|$ is the magnitude of the reciprocal variable to $\mathbf{r}$. The d-dimensional Fourier transform of any integrable radial function $f(r)$ is

$$\tilde{f}(k) = (2\pi)^{d/2} \int_0^\infty r^{d-1} f(r) \frac{J_{(d/2)-1}(kr)}{(kr)^{(d/2)-1}} dr, \tag{5}$$

and the inverse transform of $\tilde{f}(k)$ is given by

$$f(r) = \frac{1}{(2\pi)^{d/2}} \int_0^\infty k^{d-1} \tilde{f}(k) \frac{J_{(d/2)-1}(kr)}{(kr)^{(d/2)-1}} dk. \tag{6}$$

Here k is the wavenumber (reciprocal variable) and $J_v(x)$ is the Bessel function of order v.

A special point process of central interest in this paper is a lattice. A *lattice* Λ in $\mathbb{R}^d$ is a subgroup consisting of the integer linear combinations of vectors that constitute a basis for $\mathbb{R}^d$, *i.e.*, the lattice vectors $\mathbf{p}$; see ref. 13 for details. In a lattice Λ, the space $\mathbb{R}^d$ can be geometrically divided into identical regions F called *fundamental cells*, each of which corresponds to just one point as in Fig. 1. In the physical sciences, a lattice is equivalent to a Bravais lattice. Unless otherwise stated, for this situation we will use the term lattice. Every lattice has a dual (or reciprocal) lattice Λ^* in which the sites of that lattice are specified by the dual (reciprocal) lattice vectors $\mathbf{q} \cdot \mathbf{p} = 2\pi m$, where $m = \pm 1, \pm 2, \pm 3 \ldots$. The dual fundamental cell F^* has volume $v_F^* = (2\pi)^d/v_F$, where v_F is the volume of the fundamental cell of the original lattice Λ, implying that the respective densities ρ and ρ^* of the real and dual lattices are related by $\rho\rho^* = 1/(2\pi)^d$. A *periodic* point process, or non-Bravais lattice, is a more general notion than a lattice because it is is obtained by placing a fixed configuration of N points (where $N \geq 1$) within one fundamental cell of a lattice Λ, which is then periodically replicated (see Fig. 1). Thus, the point process is still periodic under translations by Λ, but the N points can occur anywhere in the chosen fundamental cell. Although generally a non-Bravais lattice does not have a dual, certain periodic point patterns are known to possess *formally dual* non-Bravais lattices. Roughly speaking, two non-Bravais
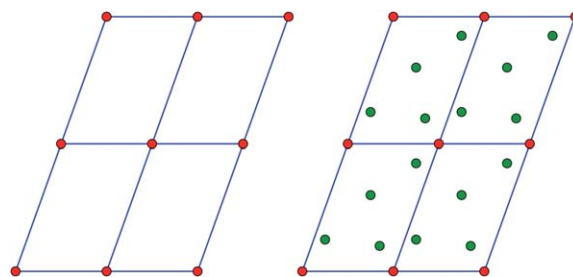

Fig. 1 Left panel: Portion of a Bravais lattice with one particle per fundamental cell. Particles are situated on the vertices of the four rhombic fundamental cells shown. Right panel: Portion of a periodic non-Bravais lattice with five particles per fundamental cell. In general, the non-Bravais lattice can have an arbitrary number of particles, each located at a different position, per fundamental cell.

lattices are formal duals of each other if their average pair sums (total energies per particle) obey the same relationship as Poisson summation for Bravais lattices for all admissible pair interactions; for further details, the reader is referred to ref. 15.

III. Duality relations

A. Pair potentials

For a configuration $\mathbf{r}^N \equiv \mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N$ of $N \gg 1$ particles in a bounded volume $V \subset \mathbb{R}^d$ with stable pairwise interactions, the many-body function

$$U(\mathbf{r}^N) = \frac{1}{N} \sum_{i=1, j=1} v(r_{ij}), \qquad (7)$$

is twice the total potential energy per particle [plus the "self-energy" $v(0)$], where $v(r_{ij})$ is a radial pair potential function and $r_{ij} = |\mathbf{r}_j - \mathbf{r}_i|$. A pair interaction $v(r)$ is stable provided that

$$\frac{1}{N} \sum_{i=1}^{N} \sum_{j=1}^{N} v(r_{ij}) \geq 0 \qquad (8)$$

for all $N \geq 1$ and all $\mathbf{r}^N \in \mathbb{R}^d$. A nonnegative Fourier transform $\tilde{v}(k)$ implies stability, but this is a stronger condition than the former.[16] A *classical ground-state* configuration (structure) within V is one that minimizes $U(\mathbf{r}^N)$. Since we will allow for disordered ground states, then we consider the general ensemble setting that enables us to treat both disordered as well as ordered configurations. The *ensemble average* of U for a statistically homogeneous and isotropic system in the thermodynamic limit is given by

$$\langle U(\mathbf{r}^N) \rangle = v(r = 0) + \rho \int_{\mathbb{R}^d} v(r) g_2(r) d\mathbf{r}, \qquad (9)$$

where $\rho = \lim_{N \to \infty, V \to \infty} N/V$ is the number density and $g_2(r)$ is the pair correlation function. In what follows, we consider those stable radial pair potentials $v(r)$ that are bounded and absolutely integrable. We call such functions *admissible* pair potentials. Therefore, the corresponding Fourier transform $\tilde{v}(k)$ exists, which we also take to be admissible, and

$$\langle U(\mathbf{r}^N) \rangle = v(r = 0) + \rho \tilde{v}(k = 0) + \rho \int_{\mathbb{R}^d} v(r) h(r) d\mathbf{r} \qquad (10)$$

Lemma. For *any ergodic configuration* in $\mathbb{R}^d$, the following duality relation holds:

$$\int_{\mathbb{R}^d} v(r) h(r) d\mathbf{r} = \frac{1}{(2\pi)^d} \int_{\mathbb{R}^d} \tilde{v}(k) \tilde{h}(k) d\mathbf{k} \qquad (11)$$

If such a configuration is a ground state, then the left and right sides of (11) are *minimized*.

Proof: We assume *ergodicity*, *i.e.*, the macroscopic properties of any single configuration in the thermodynamic limit $N, V \to +\infty$ with $\rho = N/V = $ constant are equal to their ensemble-average counterparts. The identity (11) follows from Plancherel's theorem, assuming that $\tilde{h}(k)$ exists. It follows from (10) and (11) that both sides of (11) are minimized for any ground-state structure, although the duality relation (11) applies to general (*i.e.*, non-ground-state) structures.

Remarks:

1. The general duality relation (11) was apparently not noticed or exploited before its presentation in ref. 7, although it was used for a specific pair interaction in ref. 13. The reason for this

perhaps is due to the fact that one is commonly interested in the total energy or, equivalently, the integral of (9) for which Plancherel's theorem cannot be applied because the Fourier transform of $g_2(r)$ does not exist.

2. It is important to recognize that whereas $h(r)$ always characterizes a point process,[13] its Fourier transform $\tilde{h}(k)$ is generally not the total correlation function of a point process in reciprocal space. It is when $h(r)$ characterizes a Bravais lattice Λ (a special point process) that $\tilde{h}(k)$ is the total correlation function of a point process, namely the reciprocal Bravais lattice Λ^*. One can also generalize this statement to include periodic non-Bravais lattices that possess formal duals.

3. The ensemble-averaged structure factor is related to the collective density variable $\sum_{j=1}^{N} \exp(i\mathbf{k} \cdot \mathbf{r}_j)$ *via* the expression $\lim_{N \to \infty} \left\langle \frac{1}{N} \left| \sum_{j=1}^{N} \exp(i\mathbf{k} \cdot \mathbf{r}_j) \right|^2 \right\rangle = (2\pi)^d \rho \delta(\mathbf{k}) + S(k)$, where $S(k)$ is defined in (4).

4. On account of the "uncertainty principle" for Fourier pairs, the duality relation (11) provides a computationally fast and efficient way of computing energies per particle of configurations for a non-localized (long-ranged) potential, say $v(r)$, by evaluating the equivalent integral in reciprocal space for the corresponding localized (compact) dual potential $\tilde{v}(k)$.[7]

Theorem 1. If an admissible pair potential $v(r)$ has a Bravais lattice Λ ground-state structure at number density ρ, then we have the following duality relation for the minimum U_{min} of U:

$$v(r = 0) + \sum_{\mathbf{r} \in \Lambda}' v(r) = \rho \tilde{v}(k = 0) + \rho \sum_{\mathbf{k} \in \Lambda^*}' \tilde{v}(k), \qquad (12)$$

where the prime on the sum denotes that the zero vector should be omitted, Λ^* denotes the reciprocal Bravais lattice,[†] and $\tilde{v}(k)$ is the dual pair potential, which automatically satisfies the stability condition, and therefore is admissible. Moreover, the minimum U_{min} of U for any ground-state structure of the dual potential $\tilde{v}(k)$, is bounded from above by the corresponding real-space *minimized* quantity U_{min} or, equivalently, the right side of (12), *i.e.*,

$$\tilde{U}_{min} \leq U_{min} = \rho \tilde{v}(k = 0) + \rho \sum_{\mathbf{k} \in \Lambda^*}' \tilde{v}(k). \qquad (13)$$

Whenever the reciprocal lattice Λ^* at *reciprocal lattice density* $\rho^* = \rho^{-1}(2\pi)^{-d}$ is a ground state of $\tilde{v}(k)$, the inequality in (13) becomes an equality. On the other hand, if an admissible dual potential $\tilde{v}(k)$ has a Bravais lattice Λ^* at number density ρ^*, then

$$U_{min} \leq \tilde{U}_{min} = \rho^* v(r = 0) + \rho^* \sum_{\mathbf{r} \in \Lambda}' v(r), \qquad (14)$$

where equality is achieved when the real-space ground state is the lattice Λ reciprocal to Λ^*.

Proof: We provide formal details of the proof here and refer the reader to ref. 7 for the full proof. Both the total correlation function and its Fourier transform are realizable for a Bravais lattice and its dual; specifically,

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† Note that the duality relation (12) is an expanded form of the well-known Poisson summation formula, *i.e.*, the zero-vector contributions are explicitly displayed.

$$h(r) = \frac{1}{\rho s_1(r)} \sum_{n=1} Z_n \delta(r - r_n) - 1, \quad (15)$$

where $s_1(r)$ is the surface area of a d-dimensional sphere of radius r, Z_n is the coordination number (number of points) at the radial distance r_n, and $\delta(r)$ is a radial Dirac delta function. Substituting this relation and the corresponding expression for the dual Bravais lattice into (11) yields the duality relation (12). However, the real-space ground state energy U_{min} is generally not equal to the corresponding minimum $\tilde{U}_{min}$ associated with the ground state of the dual potential $\tilde{v}(k)$, i.e., there may be periodic structures that have lower energy than the reciprocal lattice so that $\tilde{U}_{min} \leq U_{min}$. To prove this point, notice that U for any non-Bravais lattice by definition obeys the inequality $U_{min} \leq U$. However, because the corresponding Fourier transform $\tilde{h}(k)$ of the total correlation function $h(r)$ of the non-Bravais lattice in real space generally does not correspond to a point process in reciprocal space (see Remark 2 under Lemma 1), we cannot eliminate the possibilities that there are non-Bravais lattices in reciprocal space with $\tilde{U}$ lower than U_{min}. Therefore, the inequality of (13) holds in general with equality applying whenever the ground state structure for the dual potential $\tilde{v}(k)$ is the Bravais lattice Λ^* at density ρ^*. Inequality (14) follows in the same manner as (13) when the ground state of the dual potential is known to be a Bravais lattice.

Remarks:

1. Whenever equality in relation (13) is achieved, then a ground state structure of the dual potential $\tilde{v}(k = r)$ evaluated at the real-space variable r is the Bravais lattice Λ^* at density $\rho^* = \rho^{-1}(2\pi)^{-d}$.

2. The zero-vector contributions on both sides of the duality relation (12) are crucial in order to establish a relationship between the real- and reciprocal-space "lattice" sums indicated therein. To emphasize this point, consider in $\mathbb{R}^3$ the well-known Yukawa (screened-Coloumb) potential $v(r) = \exp(-\kappa r)/r$, which has the dual potential $\tilde{v}(k) = (4\pi)/(\kappa^2 + k^2)$. At first glance, this potential would seem to be allowable because the real-space lattice sum, given on the left side of (12), is convergent. However, the reciprocal-space lattice sum on the right side does not converge. This nonconvergence arises because $v(r = 0)$ is unbounded. Equality of "infinities" is established, but of course this is of no practical value and is the reason why we demand that an admissible potential be bounded.

3. Can one identify specific circumstances in which the strict inequalities in (13) and (14) apply? In addition to the theorem below that provides one such affirmative answer to this question, we will also subsequently give a specific one-dimensional example with unusual properties.

Theorem 2. Suppose that for admissible potentials there exists a range of densities over which the ground states are side by side coexistence of two distinct structures whose parentage are two different Bravais lattices, then the strict inequalities in (13) and (14) apply at any density in this density-coexistence interval.

Proof: This follows immediately from the Maxwell double-tangent construction in the U–ρ^{-1} plane, which ensures that the energy per particle in the coexistence region at density ρ is lower than either of the two Bravais lattices.

The duality relations of Theorem 1 enable one to use information about ground states of short-ranged potentials to draw new conclusions about the nature of the ground states of long-ranged potentials and *vice versa*. Moreover, inequalities (13) and (14) provide a computational tool to estimate ground-state energies or eliminate candidate ground-state structures as obtained by annealing in Monte Carlo and molecular dynamics simulations. In the ensuing discussion, we extend the relations for pair potentials by presenting novel higher-order duality relations. Our results can be applied to understand the phase behaviors associated with several classes of admissible functions, including the special case of "self-similar" interactions for which the low- and high-density ground states can be rigorously related.

B. Three-body and higher-order interactions

We now extend the aforementioned analysis to establish for the first time duality relations for many-particle systems interacting *via* three-body and higher-order interactions. For simplicity of exposition, we begin with a detailed construction of the three-body duality relations and then generalize to the higher-order case.

We consider a statistically homogeneous N-particle interaction $\Phi_N(\mathbf{r}^N)$ with one-, two-, and three-body contributions v_1, $v_2(r_{ij})$, and $v_3(r_{ij}, r_{il}, r_{jl})$, respectively.

$$\Phi_N(\mathbf{r}^N) = v_1 + \frac{1}{N} \sum_{i<j} v_2(r_{ij}) + \frac{1}{N} \sum_{i<j<k} v_3(r_{ij}, r_{ik}, r_{jk}), \quad (16)$$

where v_3 is symmetric, bounded, and short-ranged. Taking the ensemble average of this function implies

$$\langle \Phi_N(\mathbf{r}^N) \rangle = v_1 + \left(\frac{\rho}{2}\right) \int_{\mathbb{R}^d} g_2(\mathbf{r}) v_2(r) d\mathbf{r} + \left(\frac{\rho^2}{6}\right) \int_{\mathbb{R}^{2d}} g_3(\mathbf{r}, \mathbf{s}) v_3(r, s, |\mathbf{r} - \mathbf{s}|) d\mathbf{r} d\mathbf{s}, \quad (17)$$

involving averages over single particles, pairs, and triads. Duality relations for the former two contributions have already been considered, and we therefore direct our attention to the last term in (17).

Since $g_3(\mathbf{r}, \mathbf{s}) \to 1$ as $|\mathbf{r}|, |\mathbf{s}|$, and $|r - s| \to \infty$, this function is generally not integrable, and we therefore introduce the associated three-body total correlation function $h_3(\mathbf{r}, \mathbf{s}) = g_3(\mathbf{r}, \mathbf{s}) - 1$. Application of a double Fourier transform and Plancherel's theorem implies the following three-body analog of the Lemma (11):

$$\int_{\mathbb{R}^{2d}} h_3(\mathbf{r}, \mathbf{s}) v_3(r, s, |\mathbf{r} - \mathbf{s}|) d\mathbf{r} d\mathbf{s} = \frac{1}{(4\pi^2)^d} \int_{\mathbb{R}^{2d}} \tilde{h}_3(\mathbf{k}, \mathbf{q}) \tilde{v}_3(\mathbf{k}, \mathbf{q}) d\mathbf{k} d\mathbf{q},$$

$$(18)$$

where

$$\tilde{f}(\mathbf{k}, \mathbf{q}) = \int_{\mathbb{R}^{2d}} \exp(-i\mathbf{k} \cdot \mathbf{r} - i\mathbf{q} \cdot \mathbf{s}) f(\mathbf{r}, \mathbf{s}) d\mathbf{r} d\mathbf{s}. \quad (19)$$

One can verify directly that the following relationship defines the three-body correlation function for any statistically homogeneous N-particle point pattern:

$$\rho^2 g_3(\mathbf{r}, \mathbf{s}) = \left\langle \frac{1}{N} \sum_{i \neq j \neq \ell} \delta(\mathbf{r} - \mathbf{r}_{ij}) \delta(\mathbf{s} - \mathbf{r}_{i\ell}) \right\rangle. \quad (20)$$

For a Bravais lattice, ergodicity should hold, and we can re-write (20) as

$$\rho^2 g_3(\mathbf{r}, \mathbf{s}) = \sum_{j \neq \ell}{}' \delta(\mathbf{r} - \mathbf{r}_j)\delta(\mathbf{s} - \mathbf{r}_\ell), \qquad (21)$$

where the set $\{r_j\}$ in the summations includes all points of the lattice excluding the origin.

The dual Bravais lattice will possess a three-particle correlation function of the form $\tilde{g}_3(\mathbf{k},\mathbf{q}) = 1 + \rho^2 \tilde{h}_3(\mathbf{k},\mathbf{q})$, where ρ is the *real space* number density. Substituting (21) and the corresponding $\tilde{g}_3$ for the dual Bravais lattice into (18) gives the following duality relation for three-particle interactions:

$$v_1 + \sum_{j \neq \ell}{}' v_3\big(r_j, r_\ell, \|\mathbf{r}_j - \mathbf{r}_\ell\|\big) = \rho^2 \tilde{v}_1 + \rho^2 \sum_{m \neq n}{}' \tilde{v}_3(\mathbf{k}_m, \mathbf{k}_n), \quad (22)$$

where we have defined $\tilde{v}_1 \equiv \tilde{v}_3(\mathbf{0},\mathbf{0})$.

The extension of this analysis to higher-order interactions is straightforward. Specifically, we consider a n-particle bounded, symmetric, and short-ranged potential $v_n(r_{12},\ldots,r_{1n})$ with a statistically homogeneous point distribution and the associated Plancherel identity

$$\int_{\mathbb{R}^{(n-1)d}} h_n(\mathbf{r}_{12}, \ldots, \mathbf{r}_{1n}) v_n(r_{12}, \ldots, r_{1n}) d\mathbf{r}_{12} \cdots d\mathbf{r}_{1n}$$
$$= \left(\frac{1}{2\pi}\right)^{(n-1)d} \int_{\mathbb{R}^{(n-1)d}} \tilde{h}_n(\mathbf{k}_1, \ldots, \mathbf{k}_{n-1})\tilde{v}_n(\mathbf{k}_1, \ldots, \mathbf{k}_{n-1}) d\mathbf{k}_1 \cdots d\mathbf{k}_{n-1}. \qquad (23)$$

The n-particle correlation function of a Bravais lattice is

$$g_n(\mathbf{r}_1, \ldots, \mathbf{r}_{n-1}) = \sum_{\{\alpha\}_{n-1}}{}' \delta(\mathbf{r}_1 - \mathbf{r}_{\alpha_1}) \cdots \delta(\mathbf{r}_{n-1} - \mathbf{r}_{\alpha_{n-1}}), \qquad (24)$$

where $\{\alpha\}_{n-1}$ denotes all sets of $n-1$ distinct vectors in a Bravais lattice, excluding the origin, and α indexes the lattice points. Using this relationship, we find the following general n-particle duality relation:

$$v_1 + \sum_{\{\alpha\}_{n-1}}{}' v_n(\{\mathbf{r}_\alpha\}) = \rho^{n-1}\left[\tilde{v}_1 + \sum_{\{\alpha\}_{n-1}}{}' \tilde{v}_n(\{\mathbf{k}_\alpha\})\right], \qquad (25)$$

where $\tilde{v}_1 \equiv \tilde{v}_n(\{\mathbf{0}\})$.

IV. Applications

A. Admissible functions with compact support

Significant attention in the literature has been given to a certain class of oscillating real-space potentials $v(r)$ corresponding to a family of Fourier transforms with compact support such that the dual potential $\tilde{v}(k)$ is positive for $0 \leq k < K$ and zero otherwise.[4,5] Sütő[5] showed that in $\mathbb{R}^3$ with $K = 1$, the long-ranged real-space potential $v(r)$ has the body-centered cubic (bcc) Bravais lattice as its unique ground state at density $\rho = 1/(8\sqrt{2}\pi^3)$. At higher densities, the ground states are degenerate such that the face-centered cubic (fcc), simple hexagonal (sh), and simple cubic (sc) lattices are ground states at and above the respective densities $1/(6\sqrt{3}\pi^3)$, $\sqrt{3}/(16\sqrt{2}\pi^3)$, and $1/(8\sqrt{2}\pi^3)$.

The duality relation (12) can be applied to the Bravais lattice ground states above to infer the ground states of real-space potentials with compact support. Specifically, the duality theorem in $\mathbb{R}^3$ and Sütő's results imply that for the real-space "square-mound" potential $v(r) = \varepsilon\Theta(D-r)$, where $\Theta(x)$ is the Heaviside step function, the fcc lattice (dual of the bcc lattice) is

the unique ground state at the density $\sqrt{2}$, and the ground states are degenerate such that the bcc, sh and sc lattices are ground states at and below the respective densities $(3\sqrt{3})/4$, $2/\sqrt{3}$, and 1 (taking $D = 1$). Specific examples of such real-space potentials, for which the ground states are not rigorously known, include the aforementioned "square-mound" potential[17] and what we call here the "overlap" potential. This latter potential corresponds to the intersection volume of two d-dimensional spheres of diameter D whose centers are separated by a distance r, divided by the volume of a sphere; see ref. 13 for an explicit relation for any d. The overlap potential, which has support in the interval $[0,D]$, remarkably arises in the consideration of the variance in the number of points within a spherical "window" of diameter D for point patterns in $\mathbb{R}^d$, and its minimizer is an open problem in number theory.[14] The d-dimensional Fourier transforms of the square mound and overlap potentials are

$$\tilde{v}(k) = \varepsilon 2^{d/2} J_{d/2}(k)/(k\pi)^{d/2} \qquad \text{(square-mound)} \qquad (26)$$

$$\tilde{v}(k) = 2^d \pi^{d/2} \Gamma(1 + d/2) J_{d/2}^2(k/2)/k^d \qquad \text{(overlap)}, \qquad (27)$$

where we have chosen $D = 1$. Fig. 2 shows the real-space and dual potentials for these examples in three dimensions.

One can rationalize the densities at which the aforementioned lattices are ground state structures by examining either the square-mound or the overlap potential. The fcc lattice is the unique ground state at the density $\sqrt{2}$ because at this value (with unit nearest-neighbor distance) and lower densities the lattice energy is zero. At a slightly higher density, each of the 12 nearest neighbors contributes an amount ε to the lattice energy. At densities lower than $\sqrt{2}$, there is an uncountably infinite number of degenerate ground states. This includes the bcc, sh and sc lattices, which are also minimum-energy configurations at and below the respective densities $(3\sqrt{3})/4$, $2/\sqrt{3}$, and 1 because those are the threshold values at which these structures have lattice energies that change discontinuously from some positive value (determined by nearest neighbors only) to zero. Moreover, any structure, periodic or not, in which the nearest-neighbor distance is greater than unity is a ground state.

However, at densities corresponding to nearest-neighbor distances that are less than unity, rigorous prediction of the possible ground-state structures is considerably more difficult. For example, it has been argued in ref. 10 that real-space potentials whose Fourier transforms oscillate about zero will
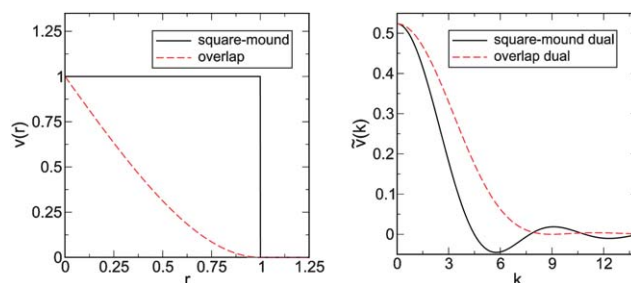

Fig. 2 Left panel: The square-mound potential $v(r) = \Theta(1-r)$ and the three-dimensional overlap potential $v(r) = \Theta(1-r)[1 - 3r/2 + r^3/2]$, where $\Theta(x)$ is the Heaviside step function. Right panel: Corresponding dual potentials $\tilde{v}(k) = \pi^{3/2} J_{3/2}(k)/(2k)^{3/2}$ (square-mound; scaled by $\pi^3/6$ for clarity) and $\tilde{v}(k) = 6\pi^2 [J_{3/2}(k/2)]^2/k^3$ (overlap).

exhibit polymorphic crystal phases in which the particles that comprise a cluster sit on top of each other. The square-mound potential is a special case of this class of potentials, and the fact that it is a simple piecewise constant function allows for a rigorous analysis of the clustered ground states for densities in which the nearest-neighbor distances are less than the distance at which the discontinuity in $v(r)$ occurs.[10]

B. Nonnegative admissible functions

The "overlap" potential discussed above also belongs to a class of admissible functions for which both $v(r)$ and $\tilde{v}(k)$ are nonnegative (*i.e.*, purely repulsive) for their entire domains. We previously introduced a pair potential corresponding to the dual of the one-dimensional overlap potential that was argued to exhibit an infinite number of structural phase transitions from Bravais to non-Bravais lattices.[7] Here we provide new numerical and analytical arguments to show that these transitions likely arise from nonanalyticity in the thermodynamic properties of the system at integer-valued densities. Additionally, we examine the three-body generalization of the so-called Gaussian core pair potential, which also belongs to this class of nonnegative admissible functions, and demonstrate the applicability of our higher-order duality relations.

1. One-dimensional overlap potential. Here we examine the one-dimensional ground-state structures associated with the dual of the so-called overlap potential

$$v(r) = \left(1 - \frac{r}{D}\right)\Theta(D - r), \qquad (28)$$

which is equal to the intersection volume, scaled by D, of two rods of radius $D/2$ with centers separated by a distance r. The dual potential is

$$\tilde{v}(k) = D\left[\frac{\sin(kD/2)}{(kD/2)}\right]^2; \qquad (29)$$

Fig. 3 shows that both potentials are bounded and repulsive. However, while the overlap potential possesses the compact support $[0,D]$, the dual potential is long-ranged with a countably infinite number of global minima determined by the zeros $k^* = 2m\pi/D$ ($m \in \mathbb{N}$) of $\sin(kD/2)$. Torquato and Stillinger have shown[14] that the unique ground state of the $d = 1$ overlap potential is the integer lattice with density $\rho = 1/D$; Theorem 1 therefore implies that the integer lattice at reciprocal density $\rho^* = D/(2\pi)$ is the unique ground state of the dual potential (29). This result intuitively corresponds to placing each point in an energy
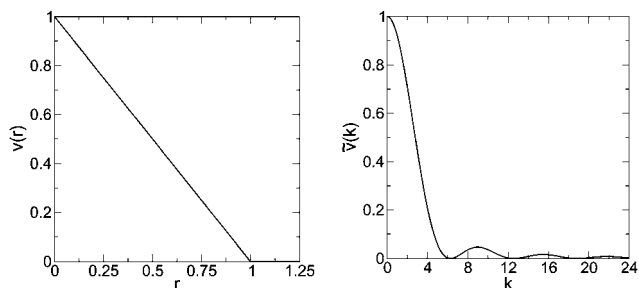

Fig. 3 Left panel: One-dimensional overlap potential $v(r) = \Theta(1 - r)(1 - r)$. Right panel: Corresponding dual potential $\tilde{v}(k) = 4\sin^2(k/2)/k^2$.

minimum of the dual potential, thereby driving the total potential energy to zero. This argument immediately implies that the integer lattice at reciprocal density $\rho^* = D/(2\pi m)$ for all $m \in \mathbb{N}$ is also a ground state of the dual potential; however, the ground states at intermediate densities are generally non-Bravais lattices and have heretofore been unexplored. Based on these observations, previous work has argued that the dual interaction (29) undergoes an infinite number of structural phase transitions from Bravais or simple non-Bravais lattices to complex non-Bravais lattices over the entire density range.[18]

We have characterized the ground states of the dual overlap potential numerically using the MINOP algorithm,[19] which applies a dogleg strategy using a gradient direction when one is far from the energy minimum, a quasi-Newton direction when one is close, and a linear combination of the two when one is at intermediate distances from a solution. The MINOP algorithm has been shown to provide more reliable results than gradient-based algorithms for similar many-body energy minimization problems.[20] For simplicity, we will for the remainder of this section describe this problem in terms of a *real space* energy minimization of the potential

$$\tilde{v}(r) = \left(\frac{\sin(\pi r)}{\pi r}\right)^2; \qquad (30)$$

i.e., we seek the ground state configurations of $\tilde{v}(r)$ (a function of real-space pair separations r instead of k) at density ρ (not ρ^*) on the line.

We fix the length L of the simulation box and use a modified version of the dual potential

$$\tilde{v}(r) = \left[\frac{\sin(\pi r N\Delta/L)}{(\pi r N\Delta/L)}\right]^2, \qquad (31)$$

where N is the number of particles. Note that L/Δ provides the unit of length for the problem, allowing us to control the density of the resulting configuration by varying Δ. However, long-range contributions to the energy per particle are particularly important in determining the ground state, meaning that any simple truncation of the pair potential within a numerical simulation can bias the observed ground-state configuration. Therefore, in order for our numerical calculations to be valid, we must be able to account for these long-range effects exactly.

To address this problem, we consider a finite configuration of N particles on the unit interval subject to periodic boundary conditions. The total energy per particle of the system (*including all periodic images of the N particles in the simulation box*) is

$$E/N = \frac{1}{2N}\sum_{i,j,n}{}' \left(\frac{\sin[N\pi\Delta(r_{ij}+n)]}{N\pi\Delta(r_{ij}+n)}\right)^2 \quad (n\in\mathbb{Z}), \qquad (32)$$

where the prime in the summation indicates that $i\neq j$ when $n = 0$. Splitting the summation over n gives

$$E/N = \frac{1}{2N}\sum_{i\neq j}\left(\frac{\sin(N\pi\Delta r_{ij})}{N\pi\Delta r_{ij}}\right)^2 + \frac{1}{2N}\sum_{i,j;n\in\mathbb{N}}\left(\frac{\sin[N\pi\Delta(n+r_{ij})]}{N\pi\Delta(n+r_{ij})}\right)^2$$
$$+ \frac{1}{2N}\sum_{i,j;n\in\mathbb{N}}\left(\frac{\sin[N\pi\Delta(n-r_{ij})]}{N\pi\Delta(n-r_{ij})}\right)^2. \qquad (33)$$

We now utilize the result

$$\sin^2[N\pi\Delta(n\pm r)] = [\sin(N\pi\Delta n)\cos(N\pi\Delta r) \\ \pm\sin(N\pi\Delta r)\cos(N\pi\Delta n)]^2 \quad (34)$$

$$= \sin^2(N\pi\Delta r) \quad (N\Delta \in \mathbb{Z}), \quad (35)$$

where the condition on $N\Delta$ stated in (35) can always be fulfilled numerically for any Δ by an appropriate choice of N. Since we are interested in the thermodynamic limit (and the intrinsic quantity E/N), this restriction places no loss of generality on our analysis. We can now evaluate the summations of n analytically using the series representation of the trigamma function[21]

$$\psi^{(1)}(1\pm r) = \sum_{n=1}^{+\infty}\frac{1}{(n\pm r)^2}, \quad (36)$$

where $\psi^{(1)}(r)$ is the *trigamma function* defined by

$$\psi^{(1)}(r) = \frac{d^2\ln\Gamma(r)}{dr^2} \quad (r\in\mathbb{C}) \quad (37)$$

with $\Gamma(r)$ the usual gamma function. We therefore have the following *effective interaction* on the N particles in the simulation box:

$$E/N = \frac{1}{2N}\sum_{i\neq j}\left(\frac{\sin(N\pi\Delta r_{ij})}{N\pi\Delta}\right)^2\left[1/r_{ij}^2 + \psi^{(1)}(1+r_{ij})\right. \\ \left. + \psi^{(1)}(1-r_{ij})\right] \quad (38)$$

$$= \frac{1}{2N}\sum_{i\neq j}\phi(r_{ij}), \quad (39)$$

which is plotted in Fig. 4.

For the case $\Delta = 1$, we have numerically verified that the the integer lattice is the unique ground state (up to translation) of the dual potential; indeed, direct calculation shows that the integer lattice minimizes the potential energy (39) for all $\Delta \in \mathbb{N}$ as expected from the arguments above. However, we have also identified degenerate ground states that are non-Bravais lattices; these systems are shown in Fig. 5. Our results suggest that for $\Delta > 1$ the ground states are complex superpositions of Bravais lattices with a minimum inter-particle spacing determined by Δ. No phase transitions are observed in this density range owing to the
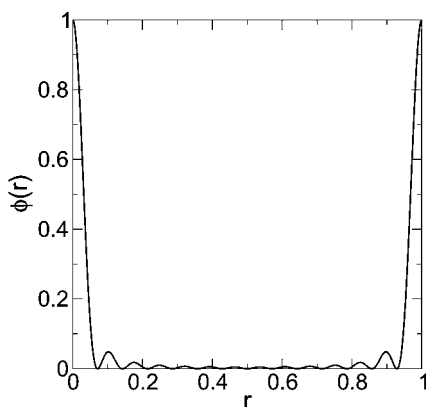

Fig. 4 Effective interaction $\phi(r)$ [*cf.* (39)] for a configuration of particles in the unit interval subject to periodic boundary conditions. The corresponding density of the dual overlap potential $v(r)$ in (30) is $\rho = 9/2$.

high degeneracy of the ground state. We remark that although the integer lattice is a ground state for any $\Delta \in \mathbb{N}$, it is never observed in our numerical simulations because the energy landscape possesses a large number of global minima. Furthermore, although the ground states for integral and non-integral values of Δ are visually similar, we emphasize that the integer lattice is never a ground-state candidate for $\Delta \notin \mathbb{N}$.

For $\Delta<1$ (density $\rho>1$), the energy per particle can no longer be driven to its global minimum of zero. Indeed, for densities $\rho \in (1,2)$, our numerical calculations *always* find the "compressed" integer lattice with spacing Δ as the ground-state structure, implying that the ground state of the dual overlap potential is unique in this density range. However, upon reaching $\rho = 2$, we are able to identify *via* numerical simulations additional candidate ground states with energy differing at most by 10^{-12} of the energy of the compressed integer lattice. Indeed, our calculations never converge to the compressed integer lattice for $\rho\geq2$, suggesting that a significant number of degenerate ground-state structures exist at these densities.

Our numerical results suggest an exact approach to characterizing the ground states of the dual potential (30). To facilitate the approach to the thermodynamic limit, we first examine a compact subset of $\mathbb{R}$ subject to periodic boundary conditions. The entropy (configurational degeneracy) of this system for $\rho \leq 1$ can be determined by relating the problem to the classic model of distributing N balls into $M \geq N$ jars such that no more than one ball occupies each jar (Fermi–Dirac statistics). Specifically, choosing the parameter $\Delta\geq1$ in (31) is equivalent to choosing a density $\rho = 1/\Delta\leq1$ in the general problem (30). Therefore, for any $\Delta\geq1$, there are $M = \Delta N$ "jars" for the N particles ("balls"). Assuming that the particles are indistinguishable, the number of distinct ways of distributing the particles into the M potential energy minima is the binomial coefficient $\binom{M}{N} = \binom{\Delta N}{N}$. For N large (approaching the thermodynamic limit), Stirling's formula implies that the entropy S is

$$S = M\ln\left(\frac{M}{M-N}\right) + N\ln\left(\frac{M-N}{N}\right) \quad (40)$$

$$S = \Delta N\ln\left(\frac{\Delta}{\Delta-1}\right) + N\ln(\Delta-1), \quad (41)$$

where we have chosen units with $k_B = 1$. Rearranging terms and substituting $\Delta = 1/\rho$ for the density, we find

$$S/N = \rho^{-1}\ln(\rho^{-1})-(\rho^{-1}-1)\ln(\rho^{-1}-1), \quad (42)$$

which is fixed in the thermodynamic limit and is plotted in Fig. 6. Note that $S/N\searrow0$ as $\rho\nearrow1$, which is expected from the observation that the integer lattice is the unique ground state at unit density. This unusual residual entropy reflects the increasing degeneracy of the ground state with decreasing density, corresponding to an increasing number of countable coexisting ground-state structures as seen in our numerical energy minimizations.

For $\rho > 1$, determination of the ground states of the dual potential (30) is nontrivial since it is no longer possible to distribute all of the particles into potential energy wells.
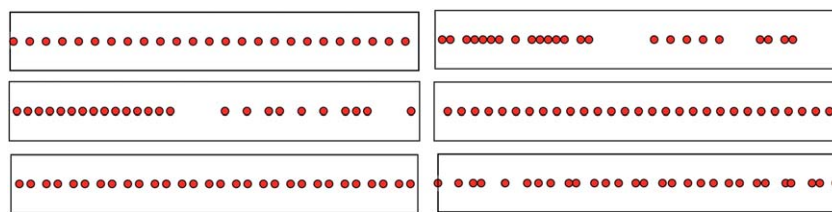
Fig. 5 Illustrative portions of numerically-determined ground state configurations of the dual potential $\tilde{v}(r)$ (30) at densities $\rho = 1$ (upper left), $\rho = 1/2$ (upper right), $\rho = 2/3$ (center left), $\rho = 3/2$ (center right), $\rho = 2$ (lower left), and $\rho = 5/2$ (lower right). The particles have been given a small but finite size for visual clarity. The $\rho = 3/2$ configuration is a compressed integer lattice with spacing $\Delta = 2/3$, and the $\rho = 2$ configuration is a "stacked" integer lattice composed of an integer lattice and its translate. The energies of each of these configurations is within at least 10^{-12} of the true ground-state energy (51).
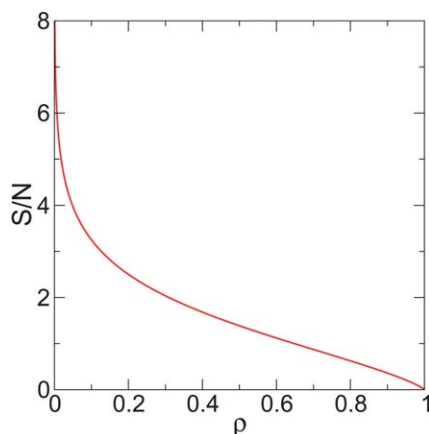

Fig. 6 Entropy per particle S/N as a function of density ρ for the dual potential $\tilde{v}(k)$ in (29) with $D = 2\pi$. Numerical evidence indicates that for $\rho \in [1,2]$ the entropy per particle is identically zero, but for higher non-integer densities, our results suggest that the entropy is nonzero. If true, this behavior implies that the system undergoes an infinite number of phase transitions as previously proposed.[7]

Therefore, Fermi–Dirac statistics are no longer applicable for the many-particle system. Nevertheless, we can make some quantitative observations concerning the ground states in this density regime. Using our duality relations, we can show analytically that the energy per particle of the "compressed" integer lattice with spacing $1/\rho < 1$ places a lower bound on the ground state energy of the dual overlap potential. Namely, we recall that twice the average energy per particle of a pair potential $v(r)$ can be written as

$$\left\langle \frac{2E}{N} \right\rangle = \left(\frac{1}{2\pi} \right)^{d} \int S(\mathbf{k})\tilde{v}(k)d\mathbf{k} + \rho\tilde{v}(0) - v(0), \quad (43)$$

where $\tilde{v}$ is the dual pair potential and $S(\mathbf{k})$ is the structure factor. Note that if the right-hand side of (43) is minimized over all $S(\mathbf{k})$ that are realizable as the pair correlation function of a point process in reciprocal space, then we obtain the following upper bound on the ground-state energy:

$$\left(\frac{2E}{N} \right)_{GS} \leq \rho \left(\frac{2E^{*}}{N} \right)_{\min S(\mathbf{k})} + \rho\tilde{v}(0) - v(0), \quad (44)$$

where E^{*} is the energy of the dual potential. For the special case that $(2E^{*}/N)_{\min S(k)} = (2E^{*}/N)_{GS}$ (such as, for example, when the

ground state is a Bravais lattice), we have the upper bound given in Section III. The strict inequality in (44) will hold when the ground-state configuration of the real-space potential possesses a structure factor that is not itself the pair correlation function of a point pattern in reciprocal space.

To obtain a lower bound on the ground-state energy, we utilize the trivial inequality

$$S(\mathbf{k}) \geq 0, \quad (45)$$

which implies that for any pair interaction with a *nonnegative* pair potential

$$\left(\frac{2E}{N} \right)_{GS} \geq \rho\tilde{v}(0) - v(0). \quad (46)$$

Note that (46) will apply for *any* many-particle configuration, including the ground state.

If we consider the dual overlap potential

$$v(r) = \left(\frac{\sin(\pi r)}{\pi r} \right)^{2}, \quad (47)$$

the Fourier transform of which is

$$\tilde{v}(k) = [1 - k/(2\pi)]\Theta(2\pi - k), \quad (48)$$

then the lower bound (46) implies

$$\left(\frac{E}{N} \right)_{GS} \geq \frac{\rho - 1}{2} \quad (49)$$

for all $\rho \in [0, +\infty)$. Note that $\tilde{v}(k) \geq 0$ for all k as is necessary to apply the lower bound (46).

However, for $\rho \geq 1$, it is easy to verify that the "inflated" integer lattice with spacing $\Delta^{*} = 1/\rho^{*} = 2\pi\rho$ is a ground state of the overlap potential $\tilde{v}(k)$ in (48) (with zero energy per particle). The upper bound (44) then implies that

$$\left(\frac{E}{N} \right)_{GS} \leq \frac{\rho - 1}{2} \quad (\rho \geq 1), \quad (50)$$

from which, by comparison with the result (49), we can conclude

$$\left(\frac{E}{N} \right)_{GS} = \frac{\rho - 1}{2} \quad (\rho \geq 1), \quad (51)$$

corresponding to the energy per particle of the *compressed* integer lattice (in real space) with spacing $\Delta = 1/\rho$. Note that this analysis does not preclude the possibility that there exist

degenerate ground-state structures within this density range as observed in our numerical energy minimizations for $\rho \geq 2$.

This result for the ground state energy based on the compressed integer lattice should be compared with the energy per particle of a "stacked" integer lattice composed of integer lattice layers (with unit spacing), each layer translated with respect to the others within [0,1]:

$$(E/N)_{\text{stack}}(\rho) = (\alpha - 1)(\alpha - 2)\Delta/2 + (\alpha - 1)[1 - (\alpha-1)\Delta] \\ (\Delta = 1/\rho; \rho \leq \alpha; \alpha \in \text{N}). \tag{52}$$

Indeed, as Fig. 7 shows, for each density $\rho \in \text{N}$, the stacked and compressed integer lattices are degenerate. This behavior reflects the fact that at these densities integer lattice "layers" can be mechanically decoupled from each other along an energy level set. Specifically, the total energy per particle remains constant if two integer lattice layers are translated with respect to each other, meaning that the lattice layers are able to "slide" with no energy cost. However, for any density $\rho \notin \text{N}$, the compressed integer lattice always possesses a lower energy. It is interesting to note that as the density becomes large, the difference in energy between the stacked and compressed integer lattices becomes increasingly small, suggesting that these structures are degenerate as $\rho \to +\infty$. Equivalently, any local energy fluctuation at high density would be sufficient to destabilize the compressed integer lattice. In particular, we can show that it is possible to relax the stacked integer lattice *via* local perturbations of particles to obtain lower energies, meaning that it is possible (and, according to our numerical calculations, likely) at high densities that such perturbed lattices are degenerate to the compressed integer lattice.

We first consider the scenario of adding one particle to a local region, subject to periodic boundary conditions, of the integer lattice of unit spacing. Since the potential energy minima of the pair interaction (30) occur on the sites of the integer lattice, the total potential energy cannot be driven to its global minimum. Symmetry of the lattice implies that, without loss of generality, we can limit the location ξ of the particle to the interval [0,0.5]. Since the energy of the underlying integer lattice is zero and the particle, by construction, will not interact with periodic images of itself, the total potential energy of the system after addition of the particle is exactly
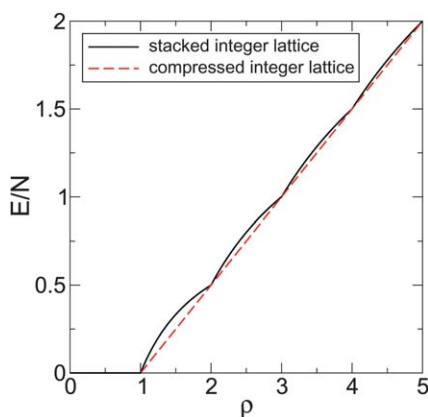
$$E = \sum_{n=0}^{+\infty} \left(\frac{\sin[\pi(\xi + n)]}{\pi(\xi + n)} \right)^2 + \sum_{n=0}^{+\infty} \left(\frac{\sin[\pi(1 - \xi + n)]}{\pi(1 - \xi + n)} \right)^2 \tag{53}$$

$$E = \sin^2(\pi\xi)[\psi^{(1)}(\xi) + \psi^{(1)}(1 - \xi)]/\pi^2, \tag{54}$$

where $\psi^{(1)}(x)$ is the trigamma function.[21] From the reflection property of the trigamma function,[21] the latter expression is exactly equal to unity [= $\tilde{v}(0)$] for any value of the parameter ξ.

Determination of the ground state then depends on "relaxing" the system by making a small perturbation $\gamma \leq 1$ in the underlying integer lattice (see Fig. 8). The energy E' of this perturbed system is then parametrized by the displacements γ and ξ as in Fig. 8 and is given by

$$E' = \sum_{n=0}^{+\infty} \left(\frac{\sin[\pi(1 - \gamma + n)]}{\pi(1 - \gamma + n)} \right)^2 + \sum_{n=0}^{+\infty} \left(\frac{\sin[\pi(1 + \gamma + n)]}{\pi(1 + \gamma + n)} \right)^2 \\ + \sum_{n=0}^{+\infty} \left(\frac{\sin[\pi(1 - \xi + n)]}{\pi(1 - \xi + n)} \right)^2 + \sum_{n=0}^{+\infty} \left(\frac{\sin[\pi(1 + \xi + n)]}{\pi(1 + \xi + n)} \right)^2 \\ + \left(\frac{\sin[\pi(\gamma + \xi + n)]}{\pi(\gamma + \xi + n)} \right)^2 \tag{55}$$

$$= 2 + \tilde{v}(\gamma + \xi) - \tilde{v}(\gamma) - \tilde{v}(\xi), \tag{56}$$

where we have utilized reflection and recurrence relations for the polygamma function[21] with $\tilde{v}(r)$ given by (30). Fig. 8 illustrates that E' possesses a unique minimum value $E'_{min} = 0.777216$ at $(\gamma, \xi) = (0.279376, 0.279376)$. A similar mechanism to the one we describe here is likely at work at high densities ($\rho \geq 2$), where our numerical calculations uncover a set of degenerate structures to the compressed integer lattice.
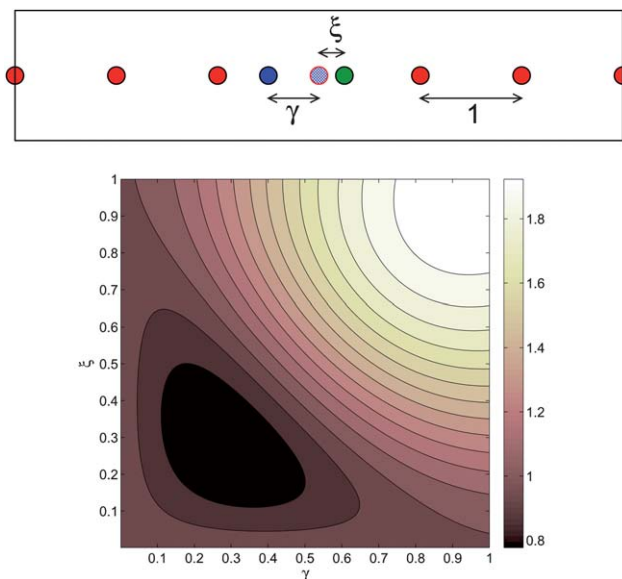

Fig. 7 Total energies per particle E/N for stacked and compressed integer lattices as functions of density ρ.

Fig. 8 Upper: Schematic illustrating the "relaxation" of the integer lattice toward the ground state of the dual potential (30) for ρ slightly greater than unity. Lower: Energy landscape associated with local perturbations of the integer lattice.

Interestingly, the long-range nature of the pair potential (30) implies that these lattices can be mechanically decoupled from each other without increasing the energy of the system. Specifically, since local perturbations in the structure affect the contributions to the total potential energy from long-range pair separations, *global* translations of the integer lattices with respect to each other can be accomplished on an energy level set as suggested in (54). Furthermore, the symmetry of this configuration implies that no simple local perturbation of the lattice structure can decrease the energy per particle, meaning that this "stacked" integer lattice and its translates within [0,1] are at least local minima of the pair interaction (30); a similar argument will hold for any $\rho \in \mathbb{N}$.

Remarks:

1. For $\rho \in [1,2)$ the compressed integer lattice appears to be the unique ground-state structure of the dual overlap potential (30). However, at higher densities, we have identified additional candidate structures that are at least numerically degenerate to the compressed integer lattice, and for any $\rho \in \mathbb{N}$ we have shown analytically that the "stacked" integer lattices are degenerate ground states. If it is true at high densities that "perturbed" stacked integer lattices are ground states, then there is a combinatorial degeneracy associated with the locations of local deformations in the underlying lattice, implying that the residual entropy is *nonanalytic* over the density range. Additionally, the system will undergo an infinite number of phase transitions as the density is increased from $\rho = 2$, thereby generalizing similar arguments for such transitions previously proposed in the literature.[7] This behavior in combination with the thermodynamic relation

$$\left(\frac{\partial(S/N)}{\partial(1/\rho)}\right)_T = \left(\frac{\partial p}{\partial T}\right)_V = \beta/\kappa_T, \tag{57}$$

where β is the thermal expansion coefficient and κ_T is the isothermal compressibility, suggests that there exist densities where the ground state exhibits negative thermal expansion as $T \to 0$.

2. One special case of the aforementioned "stacked" integer lattice configurations occurs when multiple particles occupy the same lattice sites (*i.e.*, with no translation between layers). For these "clustered" integer lattices, pair interactions are *localized* to include only those particles on the same lattice site, meaning that there are no long-range interactions for these systems. However, we have seen that the inclusion of long-range pair interactions, such as with the $\rho = 2$ integer lattice, does not affect the total energy of the system. Since relative displacements between layers are uniformly distributed on [0,1], the average displacement of 0.5 indeed corresponds to the $\rho = 2$ integer lattice.

3. Our results imply that traditional numerical methods are in general not appropriate for identifying the ground states for $\rho > 1$ since truncation of the summation (53) (*e.g.*, with the minimal image convention) breaks the translational degeneracy of the system.

4. The ground states of the overlap potential $v(r) = (1 - r) \times \Theta(1 - r)$ also exhibit rich behavior for $\rho > 1$. Since the interactions are localized to nearest neighbors, one can verify that addition of a particle to the unit density integer lattice increases the energy of the system by one unit, regardless of the position of the particle. However, unlike the dual potential (30), no local perturbation of the integer lattice can drive the system to lower energy, resulting in a large number of degenerate structures.

5. The two-dimensional ground states of the generalized dual overlap potential

$$\tilde{v}(r) = 4\pi[J_1(rD/2)/r]^2 \tag{58}$$

have also been numerically investigated;[22] the topology of the plane significantly increases the difficulty in analytically characterizing the ground-state configurations. Since the minima of the potential (58) are determined by the zeros of the Bessel function, it is not even clear that these minima coincide with the coordination shells of a periodic lattice as in one dimension. Indeed, disordered ground states for the dual overlap potential in two dimensions have been identified in certain density regimes.[22]

2. Gaussian core potential. Another interesting example of nonnegative admissible functions is the Gaussian core potential $v(r) = \varepsilon \exp[-(r/\sigma)^2]$,[23] which has been used to model interactions in polymers.[9,24] The corresponding dual potentials are self-similar Gaussian functions for any d. The potential function pairs for the case $d = 3$ with $\varepsilon = 1$ and $\sigma = 1$ are $v(r) = \exp(-r^2)$ and $\tilde{v}(k) = \pi^{3/2} \exp(-k^2/4)$. It is known from simulations[23] that at sufficiently low densities in $\mathbb{R}^3$, the fcc lattices are the ground state structures for $v(r)$. It is also known that for the range $0 \leq \rho < \pi^{-3/2}$, fcc is favored over bcc.[25] If equality in (13) is achieved for this density range, the duality theorem would imply that the bcc lattices in the range $(4\pi)^{-3/2} \leq \rho^* < \infty$ (*i.e.*, high densities) are the ground state structures for the dual potential. Lattice-sum calculations and the aforementioned simulations for the Gaussian core potential have verified that this is indeed the case, except in a narrow density interval of fcc-bcc coexistence $0.17941 \leq \rho \leq 0.17977$ around $\rho = \pi^{-3/2} \approx 0.17959$. In the coexistence interval, however, the corollary states the strict inequalities in (13) and (14) must apply. Importantly, the ground states here are not only non-Bravais lattices, they are not even periodic. The ground states are side-by-side coexistence of two macroscopic regions, but their shapes and relative orientations are expected to be rather complicated functions of density, because they depend on the surface energies of grain boundaries between the contacting crystal domains. Proposition 9.6 of ref. 26 enables us to conclude that the integer lattices are the ground states of the Gaussian core potential for all densities in one dimension. Note that in $\mathbb{R}^2$, the triangular lattices apparently are the ground states for the Gaussian core potential at all densities (even if there is no proof of such a conclusion), and therefore would not exhibit a phase transition. Similar behavior has also been observed in four and eight dimensions, where the self-dual D_4 and E_8 lattices are the apparent ground states.[15,27] Cohn, Kumar, and Schürmann have recently identified *non-Bravais lattices* in five and seven dimensions with lower ground-state energies than the densest known Bravais lattices and their duals in these dimensions.[15] Interestingly, these non-Bravais lattices, which are deformations of the D_5^+ and D_7^+ packings, possess the unusual property of *formal self-duality*, meaning that their average pair sums (total energies per particle) obey the same relation as Poisson summation for Bravais lattices for all admissible pair interactions. It is indeed an open problem to explain why formally-dual ground states exist for this pair potential.

It is also instructive to apply our higher-order duality relations (25) to the simple example of a three-body generalization of the aforementioned Gaussian-core potential. Specifically, we consider a three-body potential of the form

$$v_3(r_{12}, r_{13}, r_{23}) = \exp(-r_{12}^2 - r_{13}^2 - r_{23}^2)$$
$$= \exp[-2(r_{12}^2 + r_{13}^2 - \mathbf{r}_{12} \cdot \mathbf{r}_{13})]. \quad (59)$$

Applying a double Fourier transform to this function shows that the dual potential, given by

$$\tilde{v}_3(\mathbf{k}, \mathbf{q}) = \left(\frac{\pi^2}{3}\right)^{d/2} \exp\left[-\left(k^2 + q^2 + |\mathbf{k} - \mathbf{q}|^2\right)/12\right], \quad (60)$$

is self-similar to (59). As with the two-body version of the Gaussian-core potential, this self-similarity implies that if a Bravais lattice is the ground state of the three-body Gaussian-core interaction at low density, then its dual lattice will be the ground state at high density with the exception of a narrow interval of coexistence around the self-dual density $\rho^* = (3/\pi^2)^{d/4}$. However, we have been unable to find either numerical or analytical studies of the ground states of this higher-order interaction in the literature, and determining whether it shares ground states with its two-body counterpart is an open problem.

C. Completely monotonic admissible functions

A radial function $f(r)$ is completely monotonic if it possesses derivatives $f^{(n)}(r)$ for all $n \geq 0$ and if $(-1)^n f^{(n)}(r) \geq 0$. A radial function $f(r)$ is completely monotonic if and only if it is the Laplace transform of a finite nonnegative Borel measure μ on $[0, \infty]$, i.e., $f(r) = \int_0^\infty e^{-rt} d\mu(t)$.[28] Not all completely monotonic functions are admissible (e.g., the pure power-law potential $1/r^\gamma$ in $\mathbb{R}^d$ is inadmissible). Examples of completely monotonic admissible functions in $\mathbb{R}^d$ include $\exp(-\alpha r)$ for $\alpha > 0$ and $1/(r + \alpha)^\beta$ for $\alpha > 0$, $\beta > d$. Importantly, the Fourier transform $\tilde{f}(k)$ of a completely monotonic radial function $f(r)$ is completely monotonic in k^2.[29]

Surprisingly, the ground states of the pure exponential potential have not been investigated. Here we apply the duality relations to the real-space potential $v(r) = \exp(-r)$ in $\mathbb{R}^d$ and its corresponding dual potential $\tilde{v}(k) = c(d)/(1 + k^2)^{(d+1)/2}$ [where $c(d) = 2^d \pi^{(d-1)/2} \Gamma[(d+1)/2]$, which has a slow power-law decay of $1/k^{d+1}$ for large k. Note that the dual potential is a completely monotonic admissible function in k^2, and both $v(r)$ and $\tilde{v}(k)$ also fall within the class of nonnegative admissible functions. We have performed lattice-sum calculations for the exponential potential for a variety of Bravais and non-Bravais lattices in $\mathbb{R}^2$ and $\mathbb{R}^3$. In $\mathbb{R}^2$, we found that the triangular lattices are favored at all densities (as is true for the Gaussian core potential). If equality in (13) is achieved, then the triangular lattices are also the ground states for the slowly decaying dual potential $\tilde{v}(k) = 2\pi/(1 + k^2)^{3/2}$ at all densities. In $\mathbb{R}^3$, we found that the fcc lattices are favored at low densities ($0 \leq \rho \leq 0.017470$) and bcc lattices are favored at high densities ($0.017470 \leq \rho < \infty$). The Maxwell double-tangent construction reveals that there is a very narrow density interval $0.017469 \leq \rho \leq 0.017471$ of fcc-bcc coexistence. We see that qualitatively the exponential potential appears to behave like the Gaussian core potential. If equality in (13) applies

outside the coexistence interval, then the duality theorem would predict that the ground states of the slowly-decaying dual potential $\tilde{v}(k) = 8\pi/(1 + k^2)^2$ are the fcc lattices for $0 \leq \rho^* \leq 0.230750$ and the bcc lattice for $0.230777 \leq \rho^* < \infty$. Note that in one dimension, it also follows from the work of Cohn and Kumar[26] that since the integer lattices are the ground states of the Gaussian potential, then these unique Bravais lattices are the ground states of both the exponential potential and its dual evaluated at $k = r$ (i.e., $v(r) = 2/(1 + r^2)$).

Cohn and Kumar[26] have rigorously proved that certain configurations of points interacting with completely monotonic potentials on the surface of the unit sphere in arbitrary dimension were energy-minimizing. They also studied ways to possibly generalize their results for compact spaces to Euclidean spaces and conjectured that the densest Bravais lattices in $\mathbb{R}^d$ for the special cases $d = 2$, 8 and 24 are the unique energy-minimizing configurations for completely monotonic functions. These particular lattices are self-dual and therefore phase transitions between different lattices is not possible. Note that if the ground states for completely monotonic functions of squared distance in $\mathbb{R}^d$ (the Gaussian function being a special case) can be proved for any $d \geq 2$, it immediately follows from ref. 26 that the completely monotonic functions of distance share the same ground states. Thus, proofs for the Gaussian core potential automatically apply to the exponential potential as well as its dual (i.e., $v(r) = c(d)/(1 + r^2)^{(d+1)/2}$) because the latter is also completely monotonic in r^2.

Based upon the work of Cohn and Kumar,[26] it was conjectured that the Gaussian core potential, exponential potential, the dual of the exponential potential, and any other admissible potential function that is completely monotonic in distance or squared distance share the same ground-state structures in $\mathbb{R}^d$ for $2 \leq d \leq 8$ and $d = 24$, albeit not at the same densities.[7] Moreover, it was also conjectured for any such potential function, the ground states are the Bravais lattices corresponding to the densest known sphere packings[30] for $0 \leq \rho \leq \rho_1$ and the corresponding reciprocal Bravais lattices for $\rho_2 \leq \rho < \infty$, where ρ_1 and ρ_2 are the density limits of phase coexistence of the low- and high-density phases, respectively. In instances in which the Bravais and reciprocal lattices are self-dual ($d = 2$, 4, 8 and 24) $\rho_1 = \rho_2$, otherwise $\rho_2 > \rho_1$ (which occurs for $d = 3$, 5, 6 and 7). The second conjecture was recently shown by Cohn and Kumar to be violated for $d = 5$ and $d = 7$. Specifically, they found examples of non-Bravais lattices in five and seven dimensions related to deformations of D_5^+ (see ref. 30 for details on this structure) with low-density ground state energies in the Gaussian core potential approximately 2% lower than the densest Bravais lattices.[31] Similar results are also found at higher densities.[15] However, no counterexample for the first conjecture has been found to date, and the second conjecture is only slightly violated by the non-Bravais structures.

V. Self-dual families of pair potentials

Our discussion of the Gaussian core model above suggests that one can exactly map the energy of a lattice at density ρ to that of its dual lattice at reciprocal density ρ^* for pair potentials that are *self-similar* (defined below) under Fourier transform. However, beyond the Gaussian core potential, little is known about the thermodynamic phase properties of other types of self-similar

functions. Here we provide novel examples of self-similar potentials, including radial functions that are eigenfunctions of the Fourier transform. Only some of these results are known in the mathematics literature,[32] and this material has not previously been examined in the context of duality relations for classical ground states.

A. Eigenfunctions of the Fourier transform

Pair potentials that are eigenfunctions of the Fourier transform are unique in the context of the duality relations above since they preserve length scales for all densities; i.e., $\tilde{v}(k) = \lambda v(k)$ with no scaling factor in the argument. We therefore briefly review these eigenfunctions and the associated eigenvalues for radial Fourier transforms. In order to simply the discussion, we will adopt a unitary convention for the Fourier transform in this section

$$\widehat{f}(\mathbf{k}) = \left(\frac{1}{2\pi}\right)^{d/2} \int \exp(-i\mathbf{k}\cdot\mathbf{r})f(\mathbf{r})d\mathbf{r} \equiv \mathfrak{F}\{f\}(\mathbf{k}), \quad (61)$$

which differs from our previous usage only by a scaling factor. The slight change in notation ($\hat{f}$ instead of $\tilde{f}$) is intended to clarify which convention is being used.

The eigenfunctions of the Fourier transform for $d = 1$ can be derived from the generating function for the Hermite polynomials, which, when scaled by a Gaussian, is given by

$$\exp\left(-x^2/2 + 2tx - t^2\right) = \sum_{n=0}^{+\infty} \left(\frac{t^n}{n!}\right)\exp\left(-x^2/2\right)H_n(x). \quad (62)$$

Taking the Fourier transform of both sides, one obtains

$$\left(\frac{1}{2\pi}\right)^{1/2}\exp(-t^2)\int_{\mathbb{R}} \exp\left\{-(1/2)\left[x^2 - x(4t - 2ik)\right]\right\}dx$$
$$= \sum_n \left(\frac{t^n}{n!}\right)\mathfrak{F}\{\exp(-x^2/2)H_n(x)\}, \quad (63)$$

implying

$$\exp\left(-k^2/2\right)\sum_n \left[\frac{(-it)^n}{n!}\right]H_n(k)$$

$$= \sum_n \left(\frac{t^n}{n!}\right)\mathfrak{F}\{\exp(-x^2/2)H_n(x)\}. \quad (64)$$

By collecting powers of t in (64), we immediately conclude

$$\mathfrak{F}\{\exp(-x^2/2)H_n(x)\} = (-i)^n\exp(-k^2/2)H_n(k), \quad (65)$$

thereby identifying both the eigenfunctions and eigenvalues of the $d = 1$ Fourier transform. Note that the eigenvalues are real when n is even.

We now seek eigenfunctions of the radially-symmetric Fourier transform, defined here as

$$\widehat{f}(k) = \int_{\mathbb{R}^d} f(r)r^{d-1}\left[\frac{J_{d/2-1}(kr)}{(kr)^{d/2-1}}\right]dr \quad (66)$$

for an isotropic function $f(r)$. Direct substitution shows that $f(r) = \exp(-r^2/2)$ is an eigenfunction for all d with eigenvalue 1. Other eigenfunctions of the Fourier transform can be identified

by noting that they are also eigenfunctions of the d-dimensional Schrödinger equation for the radial harmonic oscillator

$$\left(-\frac{1}{2}\right)\left[\frac{d^2}{dr^2}\psi_n(r) + \left(\frac{d-1}{r}\right)\frac{d}{dr}\psi_n(r)\right] + \left(\frac{r^2}{2}\right)\psi_n(r) = E_n\psi_n(r), \quad (67)$$

where we have used the relation

$$\nabla^2 = \frac{d^2}{dr^2} + \left(\frac{d-1}{r}\right)\frac{d}{dr} \quad (68)$$

for radially-isotropic functions in d dimensions. The eigenvalues of the Schrödinger equation are $E_n=n + d/2$ for some $n \in \mathbb{N} \cup \{0\}$. The general solutions to (67) are then given by

$$\psi_k(r)=c_1(d)\exp(-r^2/2)L_k^{(d/2-1)}(r^2) \quad (k = n/2 \text{ for } n \text{ even}) \quad (69)$$

where $L_n^{(\alpha)}(x)$ is the associated Laguerre polynomial[21] and $c_1(d)$ is a dimension-dependent constant. Note that for $d = 1$

$$\phi_k(x) = \exp(-r^2/2)L_k^{(-1/2)}(r^2) \propto \exp(-r^2/2)H_{2k}(r), \quad (70)$$

and we recover the even $d = 1$ eigenfunctions of the harmonic oscillator.

To determine the eigenvalues of the radial Fourier transform, we note that if f is an eigenfunction, then it must be true that

$$\hat{f}(k) = cf(k) \quad (71)$$

for some eigenvalue c. However, it is also true that

$$f(k) = \widehat{\widehat{f}}(k) = c\widehat{f}(k) = c^2f(k). \quad (72)$$

Eqn (72) implies that either $c = \pm 1$ or $f(k) = 0$; for a nontrivial solution we conclude that the eigenvalues of the radially-symmetric Fourier transform are ± 1, which is in contrast to the general case on $\mathbb{R}^d$. This result is exactly consistent with the constraint that the index n of an eigenstate of the radial Schrödinger eqn (67) be even. Note that when $c = -1$, the Fourier transform changes the nature of the interaction (i.e., repulsive to attractive and vice versa).

B. Poly-Gaussian potential

The results above can be extended to include linear combinations of eigenfunctions of the Fourier transform; furthermore, we can generalize these functions to be simply self-similar under Fourier transform, meaning that length scales are not preserved by the transformation. Specifically, our interest is in functions for which:

$$\tilde{v}(k) = \lambda v(\mu k), \quad (73)$$

where λ and μ are constants.

As an example, we consider the Gaussian pair potential of the Gaussian core model

$$f(r,\sigma) = \exp(-(r/\sigma)^2). \quad (74)$$

The corresponding Fourier transform is given by

$$\tilde{f}(k,\sigma) = \int_{\mathbb{R}^3} \exp\left(-(r/\sigma)^2\right)\exp(i\mathbf{k}\cdot\mathbf{r}) = \pi^{3/2}\sigma^3\exp(-\sigma^2 k^2/4).$$
(75)

Now consider a pair potential $v(r)$ that is a linear combination of two Gaussians‡ as follows:

$$v(r) = A_1 f(r,\sigma_1) + A_2 f(r,\sigma_2).$$
(76)

The Fourier transform of (76) is

$$\tilde{v}(k) = \pi^{3/2}[A_1\sigma_1^3\exp(-\sigma_1^2 k^2/4) + A_2\sigma_2^3\exp(-\sigma_2^2 k^2/4)].$$
(77)

In order for $v(r)$ to be self-similar under Fourier transformation, the constants μ and λ of (73) must satisfy the following two equations for all x:

$$\pi^{3/2}A_1\sigma_1^3\exp(-\sigma_1^2 x^2/4) = \lambda A_2\exp(-(\mu x/\sigma_2)^2)$$
(78)

$$\pi^{3/2}A_2\sigma_2^3\exp(-\sigma_2^2 x^2/4) = \lambda A_1\exp(-(\mu x/\sigma_1)^2).$$
(79)

These equations will be satisfied by requiring

$$\sigma_2 = \frac{2\mu}{\sigma_1}, \lambda = (2\pi\mu)^{3/2}, A_2 = \frac{A_1\sigma_1^3}{(2\mu)^{3/2}},$$
(80)

leaving three independent parameters: μ, σ_1, and A_1. The example extends to any even number of Gaussian components. Let

$$v(r) = \sum_{j=1}^{2n} \exp\left(-(r/\sigma_j)^2\right),$$
(81)

where the σ_j are ordered by magnitude:

$$0 < \sigma_1 < \sigma_2 < ... \sigma_{2n} < +\infty$$
(82)

The corresponding Fourier transform is given by

$$\tilde{v}(k) = \pi^{3/2} \sum_{j=1}^{2n} A_j\sigma_j^3 \exp\left(-\sigma_j^2 k^2/4\right).$$
(83)

In order to ensure self-similarity, the terms can be paired and subject to the relations of the type (80). On account of the ordering condition (82), we pair terms with indices j and $2n - j + 1$, $1 \le j \le n$, and hence require

$$\sigma_{2n-j+1} = \frac{2\mu}{\sigma_j}, \lambda = (2\pi\mu)^{3/2}, A_{2n-j+1} = \frac{A_j\sigma_j^3}{(2\mu)^{3/2}}.$$
(84)

It is also possible to include an additional Gaussian to make an odd number in total. This additional term must effectively pair with itself, so that

$$\sigma_0 = (2\mu)^{1/2},$$
(85)

where the corresponding parameter A_0 is uncontrained and the subscript 0 refers to the "odd" term. These relations suggest an extension to the case of a *continuous* distribution of Gaussian widths as follows:

$$v(r) = \int_0^{(2\mu)^{1/2}} A(\sigma)\left[\exp\left(-\frac{r^2}{\sigma^2}\right) + \frac{\sigma^3}{(2\mu)^{3/2}}\exp\left(-\frac{\sigma^2 r^2}{4\mu^2}\right)\right]d\sigma.$$
(86)

The corresponding Fourier transform is given by

$$\tilde{v}(k) = (2\pi\mu)^{3/2} \int_0^{(2\mu)^{1/2}} A(\sigma)\left[\exp\left(-\mu^2 k^2/\sigma^2\right)\right.$$
$$\left. + \frac{\sigma^3}{(2\mu)^{3/2}}\exp\left(-\frac{\sigma^2\mu^2 k^2}{4\mu^2}\right)\right]d\sigma = \lambda v(\mu k),$$

as required for self-similarity, where $\lambda \equiv (2\pi\mu)^{3/2}$.

IV. Discussion and conclusions

In this work we have extended the results of ref. 7 by deriving new duality relations for interactions of arbitrarily high order that can be applied to help quantify and identify classical ground states for admissible potentials that arise in soft-matter systems. Our duality relations have implications for a variety of admissible potential functions, including potentials with compact support, nonnegative functions, and completely monotonic potentials. Of particular interest are the "self-similar" potentials that we have introduced, for which one can rigorously link the low- and high-densities ground state structures and energies.

We have additionally identified a set of pair potentials on the line related to the overlap function that exhibit a "stacking" phenomenon at certain densities in the ground state. This behavior leads to an unusual mechanical decoupling between layers of integer lattices due entirely to the form of the interaction. Although for $\rho \in [1,2)$ we find that the compressed integer lattice is the unique ground state of the dual overlap potential, our numerical calculations have uncovered a number of other candidate ground states at higher densities. This system should therefore exhibit an infinite number of phase transitions from Bravais to non-Bravais structures at sufficiently high densities as previously proposed in the literature[7] and likely possesses rich thermodynamic properties such as negative thermal expansion as $T \to 0$. This unusual behavior arises from nonanalyticity in the entropy of the system at integer-valued densities $\rho \ge 2$. Since overlap potentials arise in a variety of contexts, including the covering and quantizer problems[12] and the identification and design of hyperuniform point patterns,[14,34,35] further studies of these systems are warranted.

Among the examples we have studied, the completely monotonic functions offer a new category of potentials for which the ground states might be identified rigorously. In particular, we seek a proof of the conjecture that functions in this class share the same ground-state structures in $\mathbb{R}^d$ for $2 \le d \le 8$ and $d = 24$, albeit not at the same densities. Although it is true that these ground-state structures are certainly not Bravais lattices as previously suspected, no counterexample for this conjecture has been found to date. Since the five- and seven-dimensional ground state structures of the Gaussian core model proposed by Cohn, Kumar, and Schürmann[15,31] belong to a family of non-Bravais lattices possessing formal duals, our duality relations have

‡ Potentials of this form (albeit not satisfying our conditions for self-similarity) have been shown to describe the *effective* interactions between certain dendrimers.[33] The phase behavior associated with these interactions is quite different than the standard Gaussian core model.

implications even for these structures. It should also be empha-sized that the examples of admissible functions examined here are by no means complete.

In future work, we plan to explore whether analogous duality relations can be established for positive but small temperatures by studying the properties of the phonon spectra of admissible potentials. The development of such relations would provide a unique and useful guide for mapping the phase diagrams of many-particle interactions, including those functions belonging to the class of "self-similar" potentials that we have introduced here. Indeed, with the exception of the Gaussian core model,[27] little is known about the ground states and phase behaviors of self-similar functions. Since most of these potentials contain both repulsive and attractive components, these interactions have direct implications for spatially inhomogeneous solvent compo-sitions that simultaneously induce repulsion and attraction among macromolecules in solution. We expect that as the methodology continues to develop, duality relations of the type we have discussed here will play an invaluable role in under-standing these complex physical systems, including recent work on the covering and quantizer problems.[12]

Acknowledgements

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