New Duality Relations for Classical Ground States

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Abstract

We derive new duality relations that link the energy of configurations associated with a class of soft pair potentials to the corresponding energy of the dual (Fourier-transformed) potential. We apply them by showing how information about the classical ground states of short-ranged potentials can be used to draw new conclusions about the nature of the ground states of long-ranged potentials and vice versa. They also lead to bounds on the $T = 0$ system energies in density intervals of phase coexistence, the identification of a one-dimensional system that exhibits an infinite number of “phase transitions,” and a conjecture regarding the ground states of purely repulsive monotonic potentials.

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While classical ground states are readily produced by slowly freezing liquids in experiments and computer simulations, our theoretical understanding of them is far from complete. Much of the progress to rigorously identify ground states for given interactions has been for lattice models, primarily in one dimension [1]. The solutions in $d$-dimensional Euclidean space $\mathbb{R}^d$ for $d \geq 2$ are considerably more challenging. Recently, a “collective-coordinate” approach has been used to study and ascertain ground states in $\mathbb{R}^2$ and $\mathbb{R}^3$ for a class of interactions [2, 3]. A surprising conclusion of Ref. [2] is that there exist nontrivial disordered ground states without any long-range order [4], in addition to the expected periodic ones. Despite these advances, new theoretical tools are required to make further progress.

Here we derive new duality relations for a class of soft pair potentials that can be applied to classical ground states. We consider soft interactions since they are easier to treat theoretically and possess great importance in soft-matter systems, such as colloids, microemulsions, and polymers [5, 6, 7]. The duality relations link the energy of configurations for a pair potential $v(r)$ to that for the dual (Fourier-transformed) potential. Applications of the duality relations lead to some novel results.

For a configuration $r^N \equiv r_1, r_2, \ldots, r_N$ of $N \gg 1$ particles in volume $V \subset \mathbb{R}^d$ with stable pairwise interactions [9], $U(r^N) = \frac{1}{N} \sum_{i=1,j=1}^{N} v(r_{ij})$ is twice the total potential energy per particle [plus the “self-energy” $v(0)$], where $v(r)$ is a radial pair potential function and $r_{ij} = |r_j - r_i|$. A classical ground-state configuration is one that minimizes $U(r^N)$. Since we allow for disordered ground states, we consider the general ensemble setting. The ensemble average of $U$ for a statistically homogeneous and isotropic system in the thermodynamic limit is given by

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

where $\rho = \lim_{N \to \infty, V \to \infty} N/V$ is the number density and $g_2(r)$ is the pair correlation function. It is crucial to introduce the total correlation function $h(r) \equiv g_2(r) - 1$, which decays to zero for a disordered system. We consider those stable radial pair potentials $v(r)$ that are bounded and absolutely integrable and call such functions admissible. Thus, the corresponding Fourier transform $\tilde{v}(k)$ in $d$ dimensions [8] at wavenumber $k$ exists, which we also take to be admissible, and

$$\langle U(r^N) \rangle = v(r = 0) + \rho \tilde{v}(k = 0) + \rho \int_{\mathbb{R}^d} v(r) h(r) dr.$$
Lemma. For any ergodic configuration in $\mathbb{R}^d$, the following duality relation holds:

$$\int_{\mathbb{R}^d} v(r)h(r)dr = \frac{1}{(2\pi)^d} \int_{\mathbb{R}^d} \tilde{v}(k)\tilde{h}(k)dk \quad (3)$$

If such a configuration is a ground state, then left and right sides of (3) are minimized.

Proof: Identity (3) follows from Parseval’s theorem, assuming that $\tilde{h}(k)$ or the structure factor $S(k) \equiv 1 + \rho \tilde{h}(k)$ exists. From (2) and (3), we see that the both sides of (3) are minimized for any ground-state structure, although the duality relation applies to general structures [10].

Remark:

Whereas $h(r)$ always characterizes a point pattern, its Fourier transform $\tilde{h}(k)$ is generally not the total correlation function of a point pattern in reciprocal space. It is when $h(r)$ characterizes a Bravais lattice $\Lambda$ [11] that $\tilde{h}(k)$ is the total correlation function of a point pattern, namely, the reciprocal Bravais lattice $\tilde{\Lambda}$.

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

$$v(r = 0) + \sum_{r \in \Lambda}' v(r) = \rho \tilde{v}(k = 0) + \rho \sum_{k \in \tilde{\Lambda}}' \tilde{v}(k), \quad (4)$$

where the prime on the sum denotes the zero vector should be omitted, $\tilde{\Lambda}$ denotes the reciprocal Bravais lattice [12], 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 (4), i.e.,

$$\tilde{U}_{\min} \leq U_{\min} = \rho \tilde{v}(k = 0) + \rho \sum_{k \in \tilde{\Lambda}}' \tilde{v}(k). \quad (5)$$

Whenever the reciprocal lattice $\tilde{\Lambda}$ at reciprocal lattice density $\tilde{\rho} = \rho^{-1}(2\pi)^{-d}$ is a ground state of $\tilde{v}(k)$, the inequality in (5) becomes an equality. On the other hand, if an admissible dual potential $\tilde{v}(k)$ has a Bravais lattice $\tilde{\Lambda}$ at number density $\tilde{\rho}$, then

$$U_{\min} \leq \tilde{U}_{\min} = \tilde{\rho}v(r = 0) + \tilde{\rho} \sum_{r \in \Lambda}' v(r), \quad (6)$$

where equality is achieved when the real-space ground state is the lattice $\Lambda$ reciprocal to $\tilde{\Lambda}$. 

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Proof: The radially averaged total correlation function for a Bravais lattice, which we now assume to be a ground-state structure, is given by $h(r) = \frac{1}{\rho s_1(r)} \sum_{n=1} Z_n \delta(r - r_n) - 1$, 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. Substitution of this expression and the corresponding one for $\tilde{h}(k)$ into (3) yields $v(r = 0) + \sum_{n=1} Z_n v(r_n) = \rho \tilde{v}(k = 0) + \rho \sum_{n=1} \tilde{Z}_n \tilde{v}(k_n)$, where $\tilde{Z}_n$ is the coordination number in the reciprocal lattice at the radial distance $k_n$. Recognizing that $\sum_{n=1} Z_n v(r_n) = \sum'_{r \in \Lambda} v(r)$ (leading to $U_{\text{min}}$) and $\sum_{n=1} \tilde{Z}_n \tilde{v}(k_n) = \sum'_{k \in \tilde{\Lambda}} \tilde{v}(k)$ yields the duality relation (4). However, there may be non-Bravais lattice structures [11] that have lower energy than the reciprocal lattice so that $\tilde{U}_{\text{min}} \leq U_{\text{min}}$ [13]. Inequality (6) follows in the same manner as (5) when the ground state of $\tilde{v}(k)$ is known to be a Bravais lattice.

Remarks:

1. Whenever equality in relation (5) 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 $\tilde{\Lambda}$ at density $\tilde{\rho} = \rho^{-1}(2\pi)^{-d}$.

2. The zero-vector contributions on both sides of the duality relation (4) are crucial in order to establish a relationship between the real- and reciprocal-space “lattice” sums indicated therein [14].

3. We identify below specific instances in which the strict inequalities in (5) and (6) apply, including a theorem and a one-dimensional system 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 (5) and (6) apply at any density in this density-coexistence interval.

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

As we will see, the duality relations of Theorem 1 will 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 (5) and (6) provide a computational tool to estimate ground-state energies or eliminate candidate
ground-state structures as obtained from annealing simulations. We will now examine the
ground states of several classes of admissible functions.

**Admissible functions with compact support.**—Recently, the ground states of a class of
oscillating real-space potentials \( v(r) \) as defined by the family of Fourier transforms with
compact support such that \( \tilde{v}(k) \) is positive for \( 0 \leq k < K \) and zero otherwise have been
studied \([2, 3]\). Clearly, \( \tilde{v}(k) \) is admissible. Sütő \([3]\) showed that in \( \mathbb{R}^3 \) the corresponding real-
space potential \( v(r) \), which oscillates about zero, has the body-centered cubic (bcc) lattice as
its unique ground state at the real-space density \( \rho = 1/(8\sqrt{2}\pi^3) \) (with \( K = 1 \)). Moreover, he
showed that for densities greater than \( 1/(8\sqrt{2}\pi^3) \), 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 long-range behavior of the real-space oscillating potential \( v(r) \) might be regarded to be unrealistic by some. However, since all of the aforementioned ground states are Bravais
lattices, the duality relation \([4]\) can be applied here to infer the ground states of real-space
potentials with compact support. Specifically, application of the duality theorem in \( \mathbb{R}^3 \) and
Sütő’s results enables us to conclude that for the real-space potential \( v(r) \) that is positive
for \( 0 \leq r < D \) and zero otherwise, the fcc lattice (dual of the bcc lattice) is a 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 “square-mound” potential \([17]\) \( [v(r) = \epsilon > 0 \text{ for } 0 \leq r < 1 \)
and zero otherwise] and the “overlap” potential \([8]\), equal 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, and thus has support in the interval \([0, D]\) \([15]\). The \( d \)-dimensional
Fourier transforms of the square mound and overlap potentials are \( \epsilon 2^{d/2}J_{d/2}(k)/(k\pi)^{d/2} \) and
\( 2^d\pi^{d/2}\Gamma(1 + d/2)J^2_{d/2}(k/2)/k^d \), respectively, with \( D = 1 \). Figure \([\text{I}]\) shows the real-space and
dual potentials for these examples in \( \mathbb{R}^3 \). The densities at which the aforementioned lattices
are ground states are easily understood by appealing to either the square-mound or overlap
potential. The fcc lattice is a ground state at the density \( \sqrt{2} \) because at this value, where
the nearest-neighbor (NN) distance is unity, and lower densities, the energy is zero. At a
slightly higher density, each of the 12 nearest neighbors contributes an amount \( \epsilon \) to the
lattice energy. At densities lower than \( \sqrt{2} \), there are an uncountably infinite number of
degenerate ground states. This includes the bcc, sh and sc lattices, which join in as ground states 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 NN distance is greater than unity is a ground state.

However, at densities corresponding to NN distances that are less than unity, determination of the possible ground-state structures is considerably more difficult. For example, it has been argued in Ref. [6] (with good reason) that real-space potentials whose Fourier transforms oscillate about zero will 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 NN distances are less than the distance at which the discontinuity in $v(r)$ occurs [13].

Nonnegative admissible functions.– Another interesting class of admissible functions are those in which both $v(r)$ and $\tilde{v}(k)$ are nonnegative (i.e., purely repulsive) for their entire domains. The “overlap” potential discussed above is an example. Here we show that the dual $\tilde{v}(k) = 4\sin^2(kD/2)/(kD)^2$ of the overlap potential for $d = 1$ [$v(r) = 1 - r/D$ for $0 \leq r \leq D$ and zero otherwise] (see Fig. 2) exhibits rich behavior. For any density $\rho$, it

FIG. 1: Left: The localized square-mound potential [$v(r) = \epsilon = 1$ for $0 \leq r < 1$ and zero otherwise] and overlap potential [$v(r) = 1 - 3r/2 + r^3/2$ for $0 \leq r < 1$ and zero otherwise] in $\mathbb{R}^3$. Right: The delocalized dual square-mound potential $\tilde{v}(k) = \pi^3/2 J_{3/2}(k)/(2k)^{3/2}$ multiplied by $\pi^3/6$ and dual overlap potential $\tilde{v}(k) = 6\pi^2 J_{3/2}(k/2)/k^3$. 
can be shown that the unique Bravais (integer) lattice with spacing $\rho^{-1}$ is a ground-state structure \[8, 13\]. Moreover, using Theorem 1, we can show that for any $\rho = m$, where $m$ is a positive integer, the integer lattice at reciprocal density $\tilde{\rho} = (2\pi m)^{-1}$ is a ground-state structure for the dual potential $\tilde{v}(k)$; however, at non-integer density $\rho$, ground-state structures for $\tilde{v}(k)$ are generally non-Bravais lattices, establishing the strict inequality of duality relation \[5\] \[13\]. This latter result implies that for $\tilde{v}(k)$, the system undergoes an infinite number of “phase transitions” from Bravais to non-Bravais lattices over the entire density range. This one-dimensional example is interesting in its own right and further details about its ground states will be given elsewhere \[13\].

Another interesting example of nonnegative admissible functions is the Gaussian (core) potential $v(r) = \epsilon \exp[-(r/\sigma)^2]$ \[18\], which has been used to model interactions in polymers \[3\]. The dual potentials are self-similar Gaussian functions for any $d$. The potential function pairs for the case $d = 3$ with $\epsilon = 1$ and $\sigma = 1$ are $v(r) = \exp(-r^2)$ and $\tilde{v}(k) = \pi^{3/2} \exp(-k^2/4)$. Simulations \[18\] indicate that at sufficiently low densities in $\mathbb{R}^3$, the fcc lattices are the ground state structures for $v(r)$. For the range $0 \leq \rho < \pi^{-3/2}$, fcc is favored over bcc \[19\]. If equality in \(5\) is achieved for this density range, the duality theorem would imply that the bcc lattices in the range $(4\pi)^{-3/2} \leq \tilde{\rho} < \infty$ are the ground state structures for the dual potential. Previous work \[18\] has verified that this is 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, Theorem 2 states the strict
inequalities in (5) and (6) must apply. Importantly, the ground states here are not only non-Bravais lattices, they are not even periodic [16]. In \( \mathbb{R}^2 \), the triangular lattices apparently are the ground states for the Gaussian potential at all densities (but there is no proof), and therefore would not exhibit a phase transition. Proposition 9.6 of Ref. [20] enables us to conclude that the integer lattices are the ground states of the Gaussian potential for all densities in \( \mathbb{R} \).

**Completely monotonic (CM) admissible functions.**—A radial function \( f(r) \) is completely monotonic if it possesses derivatives \( f^{(n)}(r) \) for all \( n \geq 0 \) and if \( (-1)^n f^{(n)}(r) \geq 0 \). Not all CM functions are admissible (e.g., the power-law potential \( 1/r^\gamma \) in \( \mathbb{R}^d \) is inadmissible). Examples of admissible ones in \( \mathbb{R}^d \) include \( \exp(-\alpha r) \) for \( \alpha > 0 \) and \( 1/(r + \alpha)^\beta \) for \( \alpha > 0, \beta > d \).

Remarkably, the ground states of the pure exponential potential have not been studied. Here we apply the duality relations to the real-space potential \( v(r) = \exp(-r) \) in \( \mathbb{R}^d \) and its dual \( \tilde{v}(k) = c(d)/(1 + k^2)^{(d+1)/2} \) [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 \( \tilde{v}(k) \) is a CM admissible function in \( k^2 \), and both \( v(r) \) and \( \tilde{v}(k) \) are nonnegative admissible functions. We have evaluated lattice sums for the exponential potential for a variety of Bravais and periodic structures in \( \mathbb{R}^2 \) and \( \mathbb{R}^3 \). In \( \mathbb{R}^2 \), we found that the triangular lattices are favored for all \( \rho \) (as in the Gaussian case). If equality in (5) is achieved, the triangular lattices are also the ground states for the slowly decaying dual potential \( \tilde{v}(k) = 2\pi/(1 + k^2)^{3/2} \) for all \( \rho \). In \( \mathbb{R}^3 \), 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. The exponential potential appears to behave qualitatively like the Gaussian. If equality in (5) applies outside the coexistence interval, Theorem 1 would predict that the ground states of the dual potential \( \tilde{v}(k) = 8\pi/(1 + k^2)^2 \) are the fcc lattices for \( 0 \leq \tilde{\rho} \leq 0.230750 \) and the bcc lattice for \( 0.230777 \leq \tilde{\rho} < \infty \) [22].

**Conjecture.** The Gaussian potential, exponential potential, the dual of the exponential potential, and any other admissible potential function that is completely monotonic (CM) 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. For any such potential function, the ground states are the Bravais lattices corresponding to the densest known sphere packings [21] for
$0 \leq \rho \leq \rho_1$ and the reciprocal Bravais lattices for $\rho_2 \leq \rho < \infty$, where $\rho_1$ and $\rho_2$ are the density limits of phase coexistence of the low- and high-density phases, respectively. Whenever, 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 $7$).

This conjecture is bolstered by the recent work of Cohn and Kumar [20], who rigorously proved that certain configurations of points interacting with CM potentials on the surface of the unit sphere in arbitrary dimension were energy-minimizing [23].

In summary, we have derived and applied duality relations to help quantify and identify ground states for pair potentials that arise in soft-matter systems. Elsewhere, we will apply the duality relations to a broader category of functions beyond the pure Gaussians that are self-similar under Fourier transformation and will show that our formalism be extended to obtain corresponding duality relations for potential functions that also include three-body and higher-order interactions [13].

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[10] The general duality relation (3) does not seem to have been noticed or exploited before, although it was used for a specific pair interaction in Ref. [8]. On account of the “uncertainty principle” for Fourier pairs, it provides an efficient way to compute energies per particle 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 $\tilde{v}(k)$.

[11] In a Bravais lattice $\Lambda$, the space $\mathbb{R}^d$ can be geometrically divided into identical regions called fundamental cells, each of which contains one particle center.

[12] Note that the duality relation (4) is an expanded form of the well-known Poisson summation formula, i.e., the zero-vector contributions are explicitly displayed.

[13] S. Torquato and F. H. Stillinger, in preparation.

[14] To emphasize this point, Ref. [13] discusses why the Yukawa (screened-Coulomb) potential is inadmissible.

[15] The “overlap” potential arises in the consideration of the variance in the number of points within a spherical “window” of diameter $D$ for point patterns in $\mathbb{R}^d$.

[16] 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 crystal domains whose shapes and relative orientations are complicated functions of $\rho$.

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[23] If the ground states for CM functions of squared distance in $\mathbb{R}^d$ (the Gaussian function being a special case) can be proved for any $d \geq 2$, it follows from Ref. [20] that the CM functions of distance share the same ground states.