Ground states and formal duality relations in the Gaussian core model

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We study dimensional trends in ground states for soft-matter systems. Specifically, using a high-dimensional version of Parrinello-Rahman dynamics, we investigate the behavior of the Gaussian core model in up to eight dimensions. The results include unexpected geometric structures, with surprising anisotropy as well as formal duality relations. These duality relations suggest that the Gaussian core model possesses unexplored symmetries, and they have implications for a broad range of soft-core potentials.

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I. INTRODUCTION

Soft-matter systems are notoriously difficult to analyze theoretically, and much of what we know about their phase diagrams is based on numerical simulations. Even classical ground states can almost never be derived from first principles (see Refs. [1–5] for some rare exceptions). In this article, we place phenomena such as crystallization and solid-solid phase transitions in a broader context by studying dimensional trends in the Gaussian core model [6], in which particles interact via a Gaussian pair potential.

The Gaussian potential models the entropic effective interaction between the centers of mass of polymers [7], and it is one of the simplest and most elegant soft-core potentials. The behavior of the Gaussian core model in two and three dimensions is relatively well understood (see, for example, Refs. [8,9]), but in higher dimensions, it remains mysterious. Dimensions above three are an excellent test case for the study of phenomena such as decorrelation [10], and this fits into the long tradition in statistical mechanics of studying the effect of dimensionality in interacting systems, such as critical dimensions for mean-field behavior (see Section 16.7 in Ref. [11] for an overview).

Furthermore, higher dimensions play a fundamental role in information theory. Sphere packing is the low-density limiting case of the Gaussian core model, and sphere packings are error-correcting codes for a continuous communication channel. The dimension of the ambient space for the packing depends on the channel and coding method used, and it can be quite high in practice [12] (up to thousands of dimensions). Thus, coding theory is a powerful motivation for the study of the high-dimensional Gaussian core model.

Our conclusions are based on molecular dynamics simulations [13]. Such simulations are frequently used and often highly informative, but the computational difficulties are immense for many-body systems. Thanks to the curse of dimensionality [14], high-dimensional simulations typically require exponentially many particles, which severely limits the range of dimensions in which simulations are possible. To address this problem, we use a high-dimensional version of Parrinello-Rahman dynamics [15,16]. Instead of imposing periodic boundary conditions using a fixed background lattice, we dynamically update the lattice using the intrinsic geometry of the space of lattices. By increasing the adaptivity of the simulation, we are able to minimize the number of particles and avoid unnecessary computational complexity. This lets us carry out higher-dimensional simulations than were previously possible.

In this article, we carry out Parrinello-Rahman simulations of the Gaussian core model in dimensions two through eight. In addition to observing surprising geometrical phenomena such as anisotropy, we find formal duality relations between Gaussian core ground states at densities ρ and $1/\rho$. Although such duality is known between reciprocal Bravais lattices (see, for example, Ref. [17]), it rarely holds for other structures, and its occurrence here suggests a deeper, not yet understood symmetry of the Gaussian core model itself.

Our approach fits into a program pioneered by Gottwald *et al.* [18] and applied in Refs. [19,20]. They use genetic algorithms to search the space of candidate structures into which a fluid can freeze. Our goals are similar, but we make use of more analytic tools. Specifically, we compute gradients in the space of lattices, which enables us to use more powerful optimization techniques such as gradient descent or conjugate gradient.

II. FRAMEWORK

Consider a periodic configuration of particles in n-dimensional Euclidean space $\mathbb{R}^n$. Such a configuration is specified by an underlying Bravais lattice $\Lambda \subset \mathbb{R}^n$, together with a collection of translation vectors $v_1, \ldots, v_N$. In crystallographic terms, it is a lattice with basis. The particles are

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located at the points $x+v_i$ for $x \in \Lambda$ and $1 \le i \le N$.

Given a radial pair potential V, the average energy per particle is

$$\frac{1}{2N}\sum_{i=1}^{N}\sum_{j=1}^{N}\sum_{\substack{z\in\Lambda \\ z\neq 0 \text{ if } i=j}} V(|z+v_i-v_j|).$$

This quantity is the potential energy of the system, and the configuration is a classical ground state if it minimizes potential energy, even allowing Λ and N to vary but keeping the particle density fixed. [The density equals $N/\det(\Lambda)$, where $\det(\Lambda)$ is the absolute value of the determinant of a basis for Λ.] In other words, classical ground states correspond to the canonical ensemble at zero temperature.

Simulations typically fix Λ and allow $v_1,\ldots,v_N$ to vary. This amounts to using Λ to define periodic boundary conditions. The lattice Λ remains the same throughout this process, and it is often chosen to be proportional to a hypercubic lattice $\mathbb{Z}^n$ for computational simplicity. This imposes artificial structure on the system, and N must be chosen quite large to minimize the effects of this structure. For example, if one wants the lattice spacing in Λ to be an order of magnitude larger than the typical spacing between particles, then N must grow roughly like 10^n. For large n, this is clearly infeasible, and even for $n=6$, it requires careful use of all available computational improvements. Many simulations, such as those in Ref. [21], are therefore limited to roughly six dimensions.

By allowing Λ to vary, one might hope to use a much smaller value of N. In the most extreme case, one could take $N=1$ and study all Bravais lattice configurations. The naive dynamics then fail completely, because the forces in a Bravais lattice balance perfectly. Nevertheless, the potential energy varies dramatically between different Bravais lattices, with corresponding dynamics on the space of lattices. The Parrinello-Rahman method uses these dynamics. It can therefore simultaneously update the underlying Bravais lattice Λ and the particle locations $v_1,\ldots,v_N$.

Before describing the simulation results, we will give a derivation of the n-dimensional version of Parrinello-Rahman dynamics. It is equivalent to the original formulation in Refs. [15,16], except of course for the change in dimension. Instead of deriving it from a postulated Lagrangian, we show how it follows naturally from the intrinsic geometry of the space of lattices. Presenting the derivation gives us an opportunity to describe some of the computational issues that become important in higher dimensions, such as the use of lattice basis reduction algorithms.

III. GEOMETRY OF THE SPACE OF LATTICES

We will represent Bravais lattices by positive-definite, symmetric matrices. Specifically, there is a linear transformation with matrix T such that $\Lambda=T\mathbb{Z}^n$. If $v=Tw$, then the squared vector length v^tv (the exponent t denotes transpose and we use column vectors) is w^tT^tTw. Set $G=T^tT$. This Gram matrix represents the metric in coordinates in which the underlying lattice is $\mathbb{Z}^n$. Instead of fixing the metric and deforming the lattice, we will fix the lattice and deform the metric. This approach is used to define the intrinsic geometry on the space of lattices (see, for example, Ref. [22]), and it makes the formulas quite a bit simpler.

To simplify the notation, define the function f of squared distance by $f(s)=V(\sqrt{s})/2$. Furthermore, write the vectors $v_1,\ldots,v_N$ in the new coordinates as $v_i=Tu_i$. Now the potential energy of the system is

$$U(G)=\frac{1}{N}\sum_{w} f(w^tGw),$$

where we sum over all vectors w of the form u_i-u_j+x with $1\le i,j\le N$, $x\in\mathbb{Z}^n$, and $x\neq 0$ if $i=j$.

The gradient of this sum as a function of G equals the matrix

$$\nabla U(G)=\frac{1}{N}\sum_{w} f'(w^tGw)ww^t.$$

To see why, note that if we vary the i,j component G_{ij} of G while leaving all other entries fixed [and write $w=(w_1,\ldots,w_n)$], we find that

$$\frac{\partial}{\partial G_{ij}} f(w^tGw)=f'(w^tGw)w_iw_j.$$

When we update the configuration, we must fix $\det(G)$, so that the density of the system does not change [note that $\det(G)=\det(\Lambda)^2$]. However, the gradient does not respect this constraint. Instead, we must use the modified gradient $\tilde{\nabla}U(G)$ conditioned on fixing the determinant, which is computed as follows. If we define the standard inner product $\langle\cdot,\cdot\rangle$ on the space of symmetric matrices by $\langle A,B\rangle=\mathrm{tr}(AB)$, then to preserve $\det(G)$, we must remove the component of $\nabla U(G)$ in the direction of G^{-1}, because

$$\det(G+\varepsilon\,\nabla\,U(G))=\det(G)(1+\varepsilon\langle G^{-1},\nabla U(G)\rangle+O(\varepsilon^2)).$$

We define the modified gradient $\tilde{\nabla}U(G)$ by

$$\tilde{\nabla}U(G)=\nabla U(G)-\frac{\langle\nabla U(G),G^{-1}\rangle}{\langle G^{-1},G^{-1}\rangle}G^{-1},$$

so that $\langle G^{-1},\tilde{\nabla}U(G)\rangle=0$.

We could avoid this last complication by replacing the canonical ensemble with the grand canonical ensemble and controlling the particle density via the chemical potential. However, the modified gradient is not difficult to use, and our approach is convenient if one wishes to target a specific density.

IV. PARRINELLO-RAHMAN DYNAMICS

The lattice gradient has two components, one for changing G (computed above as the modified gradient) and one for changing $u_1,\ldots,u_N$. For the latter, if we write $w=u_i-u_j+x$, then we need the gradient of $f(w^tGw)$ as a function of u_i and u_j, which is easily computed as follows. With $w=u_i-u_j+x$, the gradient of $f(w^tGw)$ as a function of u_i is $2f'(w^tGw)Gw$. Thus, the u_i component of the gradient of potential energy is

the sum of $(2/N)f'(w^tGw)Gw$ over all w of the form u_i-u_j+x for some j plus the sum of $-(2/N)f'(w^tGw)Gw$ over all w of the form u_j-u_i+x for some j. The full lattice gradient is made up of both the G and the $u_1,\ldots,u_N$ components.

Parrinello-Rahman dynamics consists of using the lattice gradient to define forces on the configuration. Thus, G changes as well as $u_1,\ldots,u_N$. The simplest application is gradient descent, where we seek a local minimum for energy by following the negative gradient (or, better yet, using the conjugate gradient algorithm). Of course, one could also use the forces in the usual way to define accelerations rather than simply velocities, but we will focus on gradient descent here because of our interest in ground states.

The infinite sums in the algorithm must be truncated in practice, by summing only over the w such that w^tGw is at most some bound (chosen based on the decay rate of f). Writing $w=u_i-u_j+x$, we must enumerate all $x\in\mathbb{Z}^n$ with this property. To do so, we use the Fincke-Pohst algorithm [23], which is far more efficient than brute-force searches. By contrast, simply summing over all the vectors in a large box becomes exponentially inefficient in high dimensions. We also periodically apply the L^3 lattice basis reduction algorithm [24], which changes basis so as to keep the entries of G small, and we renormalize G to maintain a constant determinant (so that small numerical errors do not accumulate).

As the simulation progresses, the metric changes and the connection with the original coordinates is lost. Nevertheless, using the Cholesky decomposition [25], we can recover the Bravais lattice $T\mathbb{Z}^n$ with basis $Tu_1,\ldots,Tu_N$ from the matrix G and the vectors $u_1,\ldots,u_N$ by finding T such that $G=T^tT$.

Because of the need to use tools from lattice basis reduction theory and linear algebra, the Parrinello-Rahman method is not as simple to implement in high dimensions as more straightforward methods are. However, in compensation it adapts itself to the structure of the system being considered and can therefore provide improved results. Furthermore, it is compatible with other standard computational methods such as Ewald summation or fast multipole methods [13].

V. RESULTS FOR THE GAUSSIAN CORE MODEL

For the Gaussian core model, we take $V(r)=\exp(-\pi r^2)$ and hence $f(s)=\exp(-\pi s)/2$. The choice of the constant π amounts to fixing the length scale, and it is chosen to make V self-dual under the Fourier transform. Let ρ denote the particle density.

The ground states of this model have been thoroughly examined in up to three dimensions, although except in $\mathbb{R}^1$, no proof is known [5]. In $\mathbb{R}^2$, at all densities, the ground state is the triangular lattice A_2. In $\mathbb{R}^3$, the face-centered cubic lattice D_3 is optimal at low densities, and the reciprocal body-centered cubic lattice D_3^* is optimal at high densities [26]. The crossover point is at $\rho=1$, but in fact the Maxwell double-tangent construction (i.e., the convexity of potential energy as a function of $1/\rho$) leads to phase coexistence for $0.99899854\ldots\le\rho\le1.00100312\ldots$. This appears to give a complete description of the phase transition from D_3 to D_3^*.

TABLE I. Lowest known energies in dimension n when $\rho=1$. The third column specifies the putative ground state, with "pc_1" and "pc_2" standing for phase coexistence between D_3 and D_3^* and between $D_5^+(1.99750\ldots)$ and $D_5^+(0.50062\ldots)$, respectively.

n	Energy	State	n	Energy	State
1	0.04321740…	$\mathbb{Z}$	5	0.17434205…	pc_2
2	0.07979763…	A_2	6	0.19437337…	$\mathcal{P}_6(1.0525\ldots)$
3	0.11576766…	pc_1	7	0.21222702…	D_7^+
4	0.14288224…	D_4	8	0.22788144…	E_8

Little is known in higher dimensions, despite the connections with coding and information theory. Cohn and Kumar [5] conjectured that the E_8 and Leech lattices are universally optimal when $n=8$ or 24, respectively. (In other words, they are ground states for the Gaussian core model at all densities. As shown in Ref. [5], this implies optimality for many other potentials, such as all inverse power laws.) Torquato and Stillinger [17] conjectured that in at most eight dimensions, certain Bravais lattices are always optimal at sufficiently high or low densities, but their conjecture was disproved in five and seven dimensions [27]. Despite extensive exploration [28], the true ground states have remained a mystery. Because of the difficulty of simulation, previous studies have made use only of structures already known for other reasons. Comparing such structures in the Gaussian core model is of course of value, and it can sometimes lead to surprising results, but it provides little evidence as to the true ground states.

We have run numerous Parrinello-Rahman simulations with $2\le n\le8$, $1\le N\le24$ (and occasionally larger), and various densities, with the following results (see also Table I).

In two and three dimensions, we observe the previously known ground states. In four dimensions, we find the D_4 lattice at all densities, in accordance with the conjecture in Ref. [17]. Thus, it appears probable that, like E_8 and the Leech lattice, the D_4 lattice is universally optimal.

In five dimensions, we find different structures. The Λ_5^2 lattice was used in Ref. [27] to improve on Bravais lattices at low density; it consists of parallel translates of D_4, repeating with period 4. Parrinello-Rahman simulations rapidly identify and improve on this structure. It can be deformed by compressing the spacing between the parallel copies by some factor. A local minimum for energy is achieved for carefully optimized values of the compression factor, which are between 0.998749… and 1 when $\rho\le1$ and between 0.25 and 0.250312… when $\rho\ge1$. Our simulations suggest that these are the true ground states, with the exception of phase coexistence for $0.99836946\ldots\le\rho\le1.00163526\ldots$.

These structures fit into the following general family. Let

$$D_n=\{(x_1,\ldots,x_n)\in\mathbb{Z}^n:x_1+\ldots+x_n \text{ is even}\}$$

denote the checkerboard lattice in $\mathbb{R}^n$, and let D_n^+ be the union of D_n with its translation by $(1/2,1/2,\ldots,1/2)$. Let $D_n^+(\alpha)$ be D_n^+ with the last coordinate scaled by a factor of α. That is,

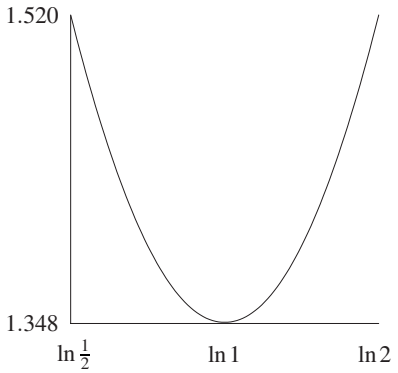

FIG. 1. A plot of $\rho^{-1/2}(2U_\rho+1)$ as a function of $\ln \rho$, where U_ρ is the minimal energy attained by the structures $D_5^+(\alpha)$ at density ρ. The reflection symmetry follows from formal duality.

$$D_n^+(\alpha) = \{(x_1, \ldots, x_{n-1}, \alpha x_n) : (x_1, \ldots, x_n) \in D_n^+\}.$$

Then $D_5^+(\alpha)$ is the deformation of Λ_5^2 with compression factor $\alpha/2$. This is not obvious, but it can be checked by a straightforward computation, and in fact it gives a substantially simpler construction of Λ_5^2 than was previously known [namely, as $D_5^+(2)$].

One noteworthy aspect of these configurations is their anisotropy. As the density increases, they experience greater compression along a distinguished axis than orthogonally to it. However, an even more surprising phenomenon is that there are formal duality relationships between these structures. Formal duality is a generalization of the relationship between a Bravais lattice and its reciprocal lattice (see Sec. VI for more details). Formal duality relates the energies at densities ρ and $1/\rho$: if E_ρ is the Gaussian core energy at density ρ for a given structure and $\tilde{E}_\rho$ is that for its formal dual, then

$$\frac{2E_\rho + 1}{2\tilde{E}_{1/\rho} + 1} = \rho.$$

More generally, formal duality relates the energy of one structure under a given pair potential to that of the formally dual structure under the Fourier transform of the potential.

A non-Bravais lattice typically has no formal dual. Thus, it is remarkable that $D_n^+(\alpha)$ is formally dual to $D_n^+(1/\alpha)$. (See Sec. VI for a proof.) Formal duality implies that the high-density ground states in $\mathbb{R}^5$ tend to $D_5^+(1/2)$, because the low-density ones tend to $D_5^+(2)=\Lambda_5^2$. Figure 1 illustrates the formal dualities within the $D_5^+(\alpha)$ family of structures.

In six dimensions, the lowest-energy states previously known were the Bravais lattice E_6 for low density and its reciprocal lattice E_6^* for high density, with a narrow region of phase coexistence in between. The lattices remain the lowest-energy states known at extreme densities, but in between them, our simulations have identified other candidate ground states. They are all deformations of the orthogonal direct sum $D_3 \oplus D_3$, together with its translates by three vectors, namely,

$$\left(\frac{1}{2},\frac{1}{2},\frac{1}{2},\frac{1}{2},\frac{1}{2},\frac{1}{2}\right),$$
$$\left(1,1,1,-\frac{1}{2},-\frac{1}{2},-\frac{1}{2}\right), \text{ and}$$
$$\left(-\frac{1}{2},-\frac{1}{2},-\frac{1}{2},1,1,1\right).$$

(These vectors are made up of holes in D_3.) Define $\mathcal{P}_6(\alpha)$ to be the structure obtained by scaling the first three coordinates by a factor of α and the last three by $1/\alpha$, so that volume is preserved. For certain values of α, these structures improve on E_6 and E_6^* for $0.25384516\ldots \le \rho \le 3.93940925\ldots$, with phase coexistence for $3.93255017\ldots \le \rho \le 3.94624440\ldots$ and for the reciprocal range of densities. Unlike $D_5^+(\alpha)$, which can be viewed as a modification of D_5^+ or Λ_5^2, the structures $\mathcal{P}_6(\alpha)$ differ more substantially from previously analyzed structures. As in five dimensions, however, there are formal duality relations. Specifically, $\mathcal{P}_6(\alpha)$ is formally dual to $\mathcal{P}_6(1/\alpha)$ for each α [of course $\mathcal{P}_6(1/\alpha)$ is isometric to $\mathcal{P}_6(\alpha)$].

In seven dimensions, we find the $D_7^+(\alpha)$ family of structures at all densities. For $0.04660088\ldots \le \rho \le 21.45881937\ldots$, the ground state seems to be D_7^+ itself (i.e., $\alpha=1$). For lower densities, we have $\alpha>1$ and for higher densities we have $\alpha<1$. Unlike the case of $\mathbb{R}^5$, there is no phase coexistence, because the optimal value of α changes continuously as a function of density. The low-density limit of $D_7^+(\alpha)$ is $D_7^+(\sqrt{2})$, which is the same as the Λ_7^3 structure studied in Ref. [27]. By formal duality, the high-density limit is $D_7^+(1/\sqrt{2})$.

Finally, in eight dimensions our simulations provide further evidence that E_8 (i.e., D_8^+) is universally optimal. Parrinello-Rahman simulations are by no means limited to eight dimensions, and in fact, we have numerical results in as many as twelve dimensions. These results, together with a more extensive analysis of the structures presented here, will appear elsewhere [29].

Within the $D_n^+(\alpha)$ family of structures for $1 \le n \le 8$, the best energy at low density (i.e., the best sphere packing) is obtained when $\alpha=\sqrt{9-n}$. For $n \le 4$, the $D_n^+(\sqrt{9-n})$ configuration is inferior to previously known sphere packings. However, for $5 \le n \le 8$ it achieves the highest sphere packing density currently known. Note that $D_6^+(\sqrt{3})$ is the Λ_6^2 packing, which has the same packing density as E_6 but is slightly inferior in the Gaussian core model at low densities. We have no conceptual explanation for why the six-dimensional behavior is subtly different from that in five, seven, or eight dimensions.

It is also interesting to examine the $\alpha=1$ case. It seems that the D_5^+ and D_6^+ structures are not local optima for the Gaussian core model at any density. By contrast, D_7^+ appears to be the ground state over a large range of densities, and D_8^+ is almost certainly the ground state at all densities. It is possible that D_9^+ is also universally optimal: so far we have not explored this case as thoroughly as those in lower dimensions, but we have not yet found any structure that beats D_9^+ at any density. We will examine this issue elsewhere [29].

Universal optimality cannot hold for D_n^+ with $n \geq 10$, because these packings are not even optimal sphere packings.

VI. FORMAL DUALITY

Recall that Poisson summation relates the sum of a function over a Bravais lattice to the sum of its Fourier transform over the reciprocal lattice. Specifically, given a sufficiently well-behaved function $f: \mathbb{R}^n \to \mathbb{R}$ (for example, a Schwartz function) and a Bravais lattice $\Lambda \subset \mathbb{R}^n$,

$$\sum_{x \in \Lambda} f(x) = \frac{1}{\mathrm{vol}(\mathbb{R}^n/\Lambda)} \sum_{y \in \Lambda^*} \hat{f}(y).$$

Here, $\mathrm{vol}(\mathbb{R}^n/\Lambda)$ denotes the volume of a fundamental domain of Λ and we normalize the Fourier transform and reciprocal lattice by

$$\hat{f}(y) = \int_{\mathbb{R}^n} f(x) e^{2\pi i \langle x,y \rangle} dx$$

and

$$\Lambda^* = \{y \in \mathbb{R}^n : \langle x,y \rangle \in \mathbb{Z} \text{ for all } x \in \Lambda\}.$$

Formal duality is a generalization of Poisson summation to certain special structures that are not Bravais lattices. To state it correctly, it is important to view the sum

$$\sum_{x \in \Lambda} f(x)$$

not simply as a sum over points in Λ, but rather as a sum over vectors between points in Λ. For a Bravais lattice these notions are exactly the same, but in more general cases they are not. (In fact, Poisson summation cannot be generalized from the first point of view [30].) For example, consider a periodic configuration given by the union of N disjoint lattice translates $\Lambda + v_1, \ldots, \Lambda + v_N$. Then the analog of summing over the lattice is

$$\frac{1}{N} \sum_{j=1}^{N} \sum_{k=1}^{N} \sum_{x \in \Lambda} f(x + v_j - v_k).$$

Call this sum the *average pair sum* of f over the configuration. It is the average over all points in the configuration of the sum of f over all vectors from it to other points. (Thus, it is independent of how the configuration is decomposed into translates of Bravais lattices.)

Suppose $\mathcal{P}$ and $\mathcal{Q}$ are particle arrangements, where $\mathcal{P}$ has particle density δ and $\mathcal{Q}$ has particle density $1/\delta$. We say $\mathcal{P}$ and $\mathcal{Q}$ are *formal duals* if for every Schwartz function f, the average pair sum of f over $\mathcal{P}$ is δ times that for $\hat{f}$ over $\mathcal{Q}$. Poisson summation shows that this definition generalizes the case of reciprocal Bravais lattices. If $\mathcal{P}$ is formally dual to itself, we call it *formally self-dual*, and if it is formally dual to an isometric copy of itself, we call it *formally isodual*. For example, the triangular lattice in the plane is not self-dual, since its reciprocal lattice is a rotated copy of itself, but it is isodual.

For any periodic configuration, one can always write the average pair sum of f in terms of $\hat{f}$, by using a generalized Poisson summation formula: for a Bravais lattice Λ and translation vector v,

$$\sum_{x \in \Lambda} f(x + v) = \frac{1}{\mathrm{vol}(\mathbb{R}^n/\Lambda)} \sum_{y \in \Lambda^*} e^{-2\pi i \langle v,y \rangle} \hat{f}(y).$$

(In fact, the right side is the Fourier expansion of the left side as a function of v that is periodic modulo Λ.) The average pair sum of f over $\Lambda + v_1, \ldots, \Lambda + v_N$ then becomes

$$\frac{N}{\mathrm{vol}(\mathbb{R}^n/\Lambda)} \sum_{y \in \Lambda^*} \hat{f}(y) \left| \frac{1}{N} \sum_{j=1}^{N} e^{2\pi i \langle v_j,y \rangle} \right|^2.$$

The factor of $N/\mathrm{vol}(\mathbb{R}^n/\Lambda)$ is the density of the configuration, so the question becomes whether the remaining sum is the average pair sum of $\hat{f}$ over some periodic structure. Except when $N=1$, it usually is not: for example, the coefficient of $\hat{f}(y)$ is generally irrational. Formal duality only arises in exceptional cases. It is not obvious when a configuration has a formal dual or, if it does have one, what the formal dual is.

Proposition 1. The D_n^+ structure is formally self-dual when n is odd or a multiple of four. When n is even but not a multiple of four, D_n^+ is formally isodual.

When n is even, D_n^+ is a Bravais lattice, whose reciprocal lattice is D_n^+ if n is a multiple of four and $D_n^+(-1)$ otherwise. When n is odd, D_n^+ is not a Bravais lattice and the duality is more subtle.

Proof. Let $v = (1/2, 1/2, \ldots, 1/2)$, so D_n^+ is the union of D_n and $D_n + v$. Then the average pair sum of f over D_n^+ is

$$\frac{1}{4} \sum_{y \in D_n^*} \hat{f}(y) |1 + e^{2\pi i \langle v,y \rangle}|^2 = \sum_{y \in D_n^*} \hat{f}(y) \frac{1 + \cos(2\pi \langle v,y \rangle)}{2}.$$

The reciprocal lattice D_n^* consists of four translates of D_n, by the vectors 0, v, $(0,0,\ldots,0,1)$, and $(1/2,1/2,\ldots,1/2,-1/2)$. In each of these four cases, the inner product $\langle v,y \rangle$ is easily understood. In the first case, it is an integer, in the second it is an integer plus $n/4$, in the third it is an integer plus 1/2, and in the fourth it is an integer plus $(n-2)/4$.

When n is odd, this yields weights of 1, 1/2, 0, and 1/2 multiplying $\hat{f}(y)$ in the four cases. Because n is odd, the vector $(1,1,\ldots,1,0)$ is in D_n and hence translating D_n by $(1/2,1/2,\ldots,1/2,-1/2)$ is equivalent to translating it by $-v$. Thus, the average pair sum for f is simply

$$\frac{1}{2} \sum_{x \in D_n} (2\hat{f}(x) + \hat{f}(x + v) + \hat{f}(x - v)),$$

which is the same as the average pair sum for $\hat{f}$ over D_n^+. It follows that D_n^+ is formally self-dual when n is odd.

An analogous computation works in the even case, or it can be verified more simply using the fact that D_n^+ is then a Bravais lattice. ■

Lemma 2. If $\mathcal{P}$ and $\mathcal{Q}$ are formally dual structures in $\mathbb{R}^n$, and $T: \mathbb{R}^n \to \mathbb{R}^n$ is an invertible linear transformation, then $T\mathcal{P}$ and $(T^t)^{-1}\mathcal{Q}$ are formally dual.

Here, T^t denotes the adjoint operator with respect to the inner product (i.e., the transposed matrix).

Proof. To compute the average pair sum for f on $T\mathcal{P}$,

simply compute it for the composition $f \circ T$ on $\mathcal{P}$. The Fourier transform of $f \circ T$ is $(\det T)^{-1}(\hat{f} \circ (T^t)^{-1})$. Now applying formal duality for $\mathcal{P}$ and $\mathcal{Q}$ completes the proof. ■

It follows immediately that $D_n^+(\alpha)$ is formally dual to $D_n^+(1/\alpha)$ when n is odd or a multiple of four: apply Lemma 2 with $\mathcal{P}=\mathcal{Q}=D_n^+$ and with T being the map that multiplies the last coordinate by α.

The formal isoduality of $\mathcal{P}_6(\alpha)$ is proved similarly. For $\alpha=1$, formal self-duality follows from a calculation much like the proof for Proposition 1. Then Lemma 2 implies that $\mathcal{P}_6(\alpha)$ is formally dual to $\mathcal{P}_6(1/\alpha)$, but of course, the two configurations are isometric.

In the literature, formal duality is usually understood to refer only to radial functions f (see, for example, p. 185 of Ref. [31]). That is a weaker condition, which depends only on the radial pair correlation functions of the structures. We have defined a stronger version of formal duality in this paper, without that restriction, because the stronger version in fact holds for the structures we find in our simulations. (Furthermore, it behaves better. For example, the proof of Lemma 2 breaks down in the radial case, because f and $f \circ T$ will generally not both be radial.) However, for the discussion in Sec. V, the pair potential is isotropic, so only the radial version of formal duality is needed. Note also that radial symmetry erases the distinction between formal self-duality and formal isoduality.

VII. CONCLUSIONS AND DISCUSSION

We have used Parrinello-Rahman dynamics to identify ground states in dimensions that were previously beyond the reach of simulation. This approach is effective because it adapts to whichever underlying Bravais lattice is most favorable. That means it probably offers little advantage in detecting disordered states or even phase coexistence, but it is appropriate whenever one anticipates a high degree of symmetry.

The formal duality relations are the most noteworthy consequence of our simulations. Such relations occur only rarely for structures other than Bravais lattices, and it is far from obvious why they arise here. A remarkable possibility is that all periodic ground states of the Gaussian core model, in any dimension, may occur in formally dual pairs. If true, this hypothesis deserves a more conceptual explanation than a case-by-case calculation, and it suggests that the model possesses deeper symmetries than are currently understood. Even if it is false, there must be a reason why formal duality arises so frequently in low dimensions.

The families of structures studied in this article have much broader applicability than just to the Gaussian core model. We believe that they will minimize many other repulsive potential functions, such as inverse power laws, although we have done relatively little experimentation in this direction.

One reason for our focus on the Gaussian core model is that it is the natural setting for studying universal optimality [5]. The known universal optima include some of the most symmetrical and beautiful geometrical configurations, with connections to many other topics such as sporadic finite simple groups and exceptional Lie algebras. Relatively few universal optima are known and any new examples are of interest. In this article, we have described simulation evidence that D_4 is likely universally optimal and that D_9^+ may be. Both cases are surprising: the analog in spherical geometry of the universal optimality of D_4 turned out to be false [32], and there have not even been any previous hints that D_9^+ might be universally optimal.

Our results also offer insight into the complexity of ground states. One measure of the complexity of a lattice is the number of Bravais lattice translates required to generate it (in crystallographic terms, the minimal size of a particle basis). Bravais lattices have complexity 1, while disordered structures can be considered to have infinite complexity. The hexagonal close-packing has complexity 2, while the structures introduced here have complexity 2 (in five and seven dimensions) and 4 (in six dimensions).

Do the complexities of ground states grow with dimension? The Torquato-Stillinger decorrelation conjecture [10] suggests that they do grow and eventually become infinite. If so, how quickly do they grow? There is a striking example in ten dimensions (the Best packing [31], which is the densest sphere packing known in $\mathbb{R}^{10}$, and which has complexity 40), but other low-dimensional ground states for repulsive potentials that have been reported in the literature typically have much smaller complexity, with the exception of phase coexistence.

In up to eight dimensions, our results for the Gaussian-core model suggest that the ground states may indeed have low complexity for most densities. The structures we have identified seem difficult to improve, even if we allow the algorithm the freedom of substantially higher complexity, and we suspect that they are the true ground states. Of course, we cannot rule out the possibility that extraordinarily high-complexity states offer tiny improvements, but we consider it unlikely.

We conclude with a computational challenge regarding simulation in high dimensions. It is undoubtedly impossible to carry out effective simulations in extremely high dimensions, but where is the threshold for feasibility? For example, is it possible in 24 dimensions? Many remarkable phenomena in mathematics and information theory (such as the Leech lattice [31]) occur there, and reliable simulation results would be very interesting.

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