Dense Regular Packings of Irregular Nonconvex Particles

Joost de Graaf,^{1,*} René van Roij,² and Marjolein Dijkstra^{1,†}

¹Soft Condensed Matter, Debye Institute for Nanomaterials Science, Utrecht University,

²Institute for Theoretical Physics, Utrecht University, Leuvenlaan 4, 3584 CE Utrecht, The Netherlands

(Received 23 June 2011; revised manuscript received 17 August 2011; published 3 October 2011)

We present a new numerical scheme to study systems of nonconvex, irregular, and punctured particles in an efficient manner. We employ this method to analyze regular packings of odd-shaped bodies, both from a nanoparticle and from a computational geometry perspective. Besides determining close-packed structures for 17 irregular shapes, we confirm several conjectures for the packings of a large set of 142 convex polyhedra and extend upon these. We also prove that we have obtained the densest packing for both rhombicuboctahedra and rhombic enneacontrahedra and we have improved upon the packing of enneagons and truncated tetrahedra.

DOI: 10.1103/PhysRevLett.107.155501

PACS numbers: 61.66.-f, 02.60.-x, 61.46.Df, 82.70.Dd

The synthesis of colloids and nanoparticles has advanced tremendously over the last decade [1-4]. Currently it is not only possible to synthesize spherical particles, but also a wide variety of convex faceted shapes, such as tetrahedra, cubes, and octahedra [1,3]. Perhaps the most remarkable advancement in synthesis techniques is the capability to create with high precision and reproducibility nonconvex, irregular, and even punctured particles, e.g., colloidal caps [4], tetrapods [2], and octapods [5]. Along with the increased availability of complex shapes, there is a concurrent increase in the study of their self-assembly into liquid [6], amorphous [7], and ordered (quasi) crystalline structures [8], as well as their material properties. Interestingly, in studying these dense configurations, materials-science research interfaces with fields as diverse as discrete geometry, number theory, and computer science [9-12].

Predictions obtained from computer simulations on the phase behavior and the self-assembled structures of these particles have been essential in guiding experimental studies and in answering fundamental mathematical questions on the packing of particles. Convex objects such as spheres [6,12] and ellipsoids [7], as well as (semi) regular [9,11,13–18] and space-filling [19] solids have been the subject of intense ongoing investigation. However, ordered structures composed of irregular nonconvex particles have hardly been studied by simulation. Implementing excluded-volume interactions for such systems imposes numerical challenges, because of the complex particle shape and the additional rotational degrees of freedom. Only recently were the first attempts made to study such systems, namely, for superdisks, superballs [20], and bowls [21].

In this Letter, we present a novel composite technique by which we numerically study the dense packings of nonconvex irregular solids, colloids, and nanoparticles. The technique consists of the following elements: the floppy box Monte Carlo method [22] (FBMC), the triangular tessellation method [23] (TT), and the triangle interference detection method [24] (TID). The choice for these individual elements is based on their successful application elsewhere. The FBMC method forms the core of the technique and is an isothermal-isobaric (NPT) ensemble Monte Carlo (MC) simulation, by which crystal structures can be predicted. The hard-particle interaction in this simulation is handled using TT, by which any shape is approximated with triangles, in tandem with TID, by which intersections of triangles are efficiently determined. Two particles overlap whenever there is an intersection between a pair of triangles in the respective TT meshes. To speed up the overlap algorithm and prevent inclusions we employed inscribed- and outscribed-sphere overlap predetection. Interior triangles can be added to a particle to further prevent inclusions, whenever the gap between a particle and its inscribed sphere is too wide. This is allowed because TID models do not have to obey topological constraints [24].

In order to demonstrate the general applicability of this method, we apply our technique to study packings of an enormous set of 142 convex polyhedra and 17 irregular particle shapes, also see the Supplemental Material [25]. This set includes a few models that contain a huge number of triangles, e.g., the colloidal cap, the Stanford bunny, and the hammerhead shark, with 3850, 3756, and 5116 triangles, respectively. All particle models have been generated using particle databases or created to resemble existing nanoparticles and colloids [2,4,5]. Systems of tessellated particles are prepared in a dilute phase. By gradually increasing the reduced pressure we are able to compress the system to a high-density crystalline state. We apply this scheme many times and select the densest packing among these. This packing is allowed to compress further to obtain a maximally compressed state, see Ref. [25] for further details. The method is typically quite fast with simulations taking minutes to hours on a modern

Princetonplein 5, 3584 CC Utrecht, The Netherlands

desktop PC [25]. Even for high-triangle models the time scales are accessible, mostly due to the advanced TID routine employed [24]. For the purpose of analyzing nanoparticle systems at lower pressures [5], soft potential terms may be added by introducing point interaction sites distributed over the particle.

It should be noted that the FBMC method is similar to the adaptive shrinking cell (ASC) method of Refs. [14,15], since both allow for a sequential search of configurational space and lattice space using a Metropolis based MC procedure. However, the FBMC method uses a lattice reduction technique [26] to avoid unphysical distortions of the unit cell, whereas the ASC algorithm employs a symmetric strain tensor. In addition, the FBMC method drives the system towards its densest configuration by employing a gradual pressure increase in an NPT-MC simulation scheme, whereas the ASC method drives compression using the negative packing fraction as the basis of its Metropolis acceptance rule. The method of Ref. [27] should also be mentioned, as a different means of determining densest packings. Of the three techniques, the FBMC method has the advantage that it can be used to explore suboptimal packings in accordance with the statistical NPT ensemble at finite pressures. The densest-packed candidate crystal structure need not be thermodynamically stable at all pressures for which the system crystallizes [5,18,19]. However, it goes beyond the scope of this Letter to determine which of the techniques is most suited to achieving densest-packed structures.

To prove the accuracy of our method, we have redetermined the lower bound ϕ_{LB} to the volume fraction of the densest packing both for the 5 Platonic and for the 13 Archimedean solids. We find excellent agreement [25] with Refs. [11,14–17]: the system was typically compressed to within 0.002 of the $\phi_{\rm LB}$ literature value. The simulations we performed yielded a very narrow distribution of crystal-structure candidates near the closest-packed configuration. The densest of these only required minimal additional compression to achieve the given densest packing fraction value $\phi_{\rm LB}$. Moreover, for truncated tetrahedra we have discovered a new crystal structure, a dimer lattice with $\phi_{LB} = 0.988...$, see Fig. 1 and Ref. [25]. This is not only mathematically interesting [28], but also relevant to the study of nanoparticle systems, since truncated tetrahedra have recently been synthesized [29].

After verifying the accuracy of our technique, we applied our method to study 17 nonconvex (irregular) shapes, some of which even contained holes, thereby going beyond existing studies. Figure 2 shows representations of the shape and predicted crystal structure for 4 different particles, see Ref. [25] for additional information. Such candidate crystal structures can be used in large simulations or theory to determine their stability using, e.g., free-energy calculations. One physical system we considered in more detail is that of the colloidal cap, see Fig. 2(a), which shows



FIG. 1 (color online). The dimer crystal structure for truncated tetrahedra that achieves a packing fraction $\phi_{LB} = 0.988...$ (a) The dimer formed by two truncated tetrahedra (blue and red; dark and light gray, respectively). (b) A piece of the crystal structure this unit cell generates; only 7 periodic images are shown. The viewpoint is such that the dimers in this crystal are pointing out of the paper, with the top triangle [as given in (a)] facing the reader.

the model that was used. The model is derived from a numerical analysis: the Hamiltonian that describes the bending and in-plane stretching elasticity terms [4,30], which govern the collapse of a shell under an external isotropic pressure, was minimized using surface evolver [31]. The cap that was obtained in this way contains 3850 triangles. We found our model to yield crystal structures similar to the ones that were obtained using a much simpler bowl-shaped model [21]: columnar, braided, and inverse braided phases [25]. Figure 2(b) shows a braided configuration. Support is thus provided for the idea that the simple bowl shape [21] captures the essential shape-related physics of these systems.

Another physical system we studied in greater detail was that of octapod-shaped nanocrystals [5] [Fig. 3(a)]. In Ref. [5] we analyzed the hierarchical self-assembly of these objects into interlocked chains and three dimensional (3D) superstructures by including attractive van der Waals interactions between the octapods. Figure 3 shows an example (simple-cubic) candidate crystal structure obtained by our method for a system of octapods with a soft interaction potential: the centroids of the tips are attracted to each other according to a short-ranged, yet deep square-well interaction. Although simulations on octapods proved technically challenging, we can conclude that such systems may be studied over a range in volume fractions and that the hard-particle overlap can be supplemented with soft interaction potentials to more accurately model the experimental system. By these two physical examples, caps and octapods, we have shown that we can study previously inaccessible nanoparticle and colloid systems, as well as approximate their shape with greater precision, if required.

In the course of our investigation we also obtained several exciting and remarkable results on the packing of



FIG. 2 (color online). Four nonconvex shapes and three of their associated crystal structures. (a) A side and bottom view of the model (3850 triangles) for a colloidal cap. (b) A piece of the double-braided structure formed by these caps. There are four particles in the unit cell, i.e., the caps form a quadrumer, each has been indicated with a different color. (c) The centrosymmetric dimer formed by two Szilassi polyhedra (blue and red; dark and light gray, respectively) that achieves the densestknown packing in relation to its unit cell (gray box). We do not show the crystal this generates, since it is difficult to make out individual particles in it even when they are color coded. (d) A centrosymmetric tetrapod dimer (red, blue) and the associated unit cell (gray box). (e) A piece of the crystal structure formed by this dimer. (f) The densest-known packing for great stellated dodecahedra, again the structure is a dimer lattice as indicated by the red and blue color coding.

faceted particles [25]. (i) We have extended the verification of Ulam's conjecture [32], which states that all convex objects pack denser than spheres, to: the first 8 regular prisms and antiprisms, the 92 Johnson solids, and the 13 Catalan solids [25]. For regular *n*-prisms and *n*-antiprisms, where *n* indicates the number of edges of the bases, the verification of Ulam's conjecture may be further extended to $n = \infty$. For regular prisms this follows from the analysis of regular *n*-gons [9,10] and the columnar way in which these prisms stack, whereas for antiprisms an outscribedcylinder estimate for ϕ_{LB} shows that it is sufficient to check up to n = 7. (ii) Our results show that there is no clear dependence between the sphericity γ , the ratio of the inscribed- and outscribed-sphere radius, and the densest packing fraction ϕ_{LB} [25]. (iii) We confirmed for 49



FIG. 3 (color online). A crystal structure obtained for octapodshaped particles, which have an additional soft interaction term. (a) The model of the octapod used in this simulation. (b) Side view of the candidate crystal structure, which is a simple cubic phase. Note that the octapods touch each other at the tips.

convex centrosymmetric particles that their densest-known lattice packing is a Bravais lattice, in accordance with the conjecture of Ref. [14]. (iv) Our data also support the conjecture [15] that convex, congruent solids without central symmetry do not necessarily pack densest in a Bravais lattice. In general noncentrosymmetric particles do not pack densest in a Bravais lattice, however, there are indications that some do. A possible example is the snub cube [Fig. 4(c)] that achieves $\phi_{LB} = 0.787...$ for N = 1particle in the unit cell (Bravais lattice). We confirmed this using an extended sample set to achieve a higher numerical accuracy [25]. This may be an indication that central symmetry is not as strong a prerequisite for 3D systems, in contrast with the result for two-dimensional (2D) packings [33]. However, our result constitutes only a possible counterexample, not a full mathematical proof. Nevertheless, the snub cube is not the only particle for which we have observed this [25], e.g., the snub



FIG. 4 (color online). Four representations of convex particles and the densest-known packing for enneagons. A rhombicuboctahedron (a) and rhombic enneaconrahedron (b). For both we have proved that its Bravais lattice packs densest. (c) The snub cube, which is not centrally symmetric, yet it achieves its densest-known packing in a Bravais lattice. (d) An enneagon. (e) The centrosymmetric-dimer (blue, red) lattice that achieves the densest packing for enneagons.

dodecahedron and the metabigyrate rhombicosidodecahedron probably achieve their densest packing for N = 1. For the snub cube and snub dodecahedron this possibility was already alluded to in Ref. [34]. (v) Many noncentrosymmetric particles, both convex and nonconvex, do form a centrosymmetric compound which achieves the densest regular packing, e.g., truncated tetrahedra [Fig. 1(a)], (anti)prisms (n < 11), Szilassi polyhedra [Fig. 2(c)], and tetrapods [Fig. 2(d)], form centrosymmetric dimers and tetrahedra form centrosymmetric quadrumers [17]. (vi) Remarkably, it appears that some noncentrosymmetric particles, e.g., the gyrate rhombicosidodecahedron and the tetrapod, have a noncentrosymmetric N = 1 packing and an N = 2 centrosymmetric-dimer packing that achieve (nearly) the same packing fraction.

For rhombicuboctahedra [RCH, Fig. 4(a)] and rhombic enneacontrahedra [RECH, Fig. 4(a)] our technique established the densest packing [25]. We found that ϕ_{LB} equals the inscribed-sphere upper bound ϕ_{UB} to the packing fraction [14] within the numerical precision. We subsequently verified this by analytic calculation: for RCH $\phi_{\rm LB} = \phi_{\rm UB} = (4/3)(4\sqrt{2} - 5)$ and for RECH $\phi_{\rm LB} = \phi_{\rm UB} = 16 - 34/\sqrt{5}$. Here we have redetermined the $\phi_{\rm UB}$ for the RCH, which was incorrectly listed as $\phi_{\text{UB}} = 1$ in Ref. [14]. Interestingly, we can now invoke the same argument as for spheres [14] and determine two additional upper bounds to the densest packing fraction, based on the largest inscribed RCH and RECH, respectively. By extension of our result for the 9-prism, we also obtained a new 2D packing [Fig. 4(e), Ref. [25]] with $\phi_{\text{LB}} = 0.901...$ for the regular Enneagon [Fig. 4(d)] that surpasses the previously obtained value of $\phi_{LB} =$ 0.897 . . . [10].

In conclusion, we have shown that regular packings of irregular, nonconvex, and punctured objects may be obtained in a rigorous way by means of our composite technique. The complex problem of the packing of such shapes has been tremendously reduced by the FBMC method and triangulation. We have predicted candidate crystal structures for (faceted) nonconvex and irregular particles, improved upon the literature values for the densest packings of a huge number of solids, and confirmed and extended upon existing conjectures on their densest packing. Moreover, we also prove that we obtained the densest packing of rhombicuboctahedra and rhombic enneacontrahedra. This is remarkable not only because it has historically [12] been exceedingly difficult to prove that the densest packing of objects is indeed achieved for a certain configuration, but also because these particles can now be used to determine new estimates for the upper bound to the maximum packing fraction for other particle shapes. Finally, we also discovered denser packings than previously obtained for noncentrosymmetric enneagons and truncated tetrahedra in centrosymmetric-dimer lattices. In addition, our method can easily be extended to study dense amorphous (granular) and quasicrystalline packings and systems of arbitrarily shaped colloids and nanoparticles with soft interaction potentials in an external field, both in and out of equilibrium. Our method thus opens the way to a more comprehensive study of the material and structure properties than has previously been considered feasible.

M. D. acknowledges financial support by a "Nederlandse Organisatie voor Wetenschappelijk Onderzoek" (NWO) Vici Grant, and R. v. R. by the Utrecht University High Potential Programme.

*j.degraaf1@uu.nl

- [†]m.dijkstra1@uu.nl
- Y. Sun and Y. Xia, Science **298**, 2176 (2002).
 L. Manna *et al.*, Nature Mater. **2**, 382 (2003).
- [3] S. C. Glotzer and M. J. Solomon, Nature Mater. 6, 557 (2007).
- [4] C. Quilliet et al., Eur. Phys. J. E 27, 13 (2008).
- [5] K. Miszta et al., Nature Mater. (in press).
- [6] J. D. Bernal and J. Mason, Nature (London) 188, 910 (1960).
- [7] A. Donev et al., Science 303, 990 (2004).
- [8] D. V. Talapin et al., Nature (London) 461, 964 (2009).
- [9] G. Kuperberg and W. Kuperberg, Discrete Comput. Geom. 5, 389 (1990).
- [10] Y. Limon Duparcmeur, A. Gervois, and J.P. Troadec, J. Phys. I (France) 5, 1539 (1995).
- [11] U. Betke and M. Henk, Comput. Geom. 16, 157 (2000).
- [12] T.C. Hales and S.P. Ferguson, Discrete Comput. Geom. 36, 1 (2006).
- [13] A. Haji-Akbari et al., Nature (London) 462, 773 (2009).
- [14] S. Torquato and Y. Jiao, Nature (London) 460, 876 (2009).
- [15] S. Torquato and Y. Jiao, Phys. Rev. E 80, 041104 (2009).
- [16] S. Torquato and Y. Jiao, Phys. Rev. E 81, 041310 (2010).
- [17] E. R. Chen, M. Engel, and S. C. Glotzer, Discrete Comput. Geom. 44, 253 (2010).
- [18] A. Haji-Akbari, M. Engel, and S.C. Glotzer, arXiv:1106.4765v2 [J. Chem. Phys. (to be published)].
- [19] U. Agarwal and F. A. Escobedo, Nature Mater. 10, 230 (2011).
- [20] Y. Jiao, F. H. Stillinger, and S. Torquato, Phys. Rev. E 79, 041309 (2009).
- [21] M. Marechal et al., Nano Lett. 10, 1907 (2010).
- [22] L. Filion et al., Phys. Rev. Lett. 103, 188302 (2009).
- [23] J. de Graaf, M. Dijkstra, and R. van Roij, Phys. Rev. E 80, 051405 (2009).
- [24] GAMMA Research Group (UNC), RAPID, http://gamma .cs.unc.edu/OBB/, 1997.
- [25] See Supplemental Material at http://link.aps.org/ supplemental/10.1103/PhysRevLett.107.155501 for details on the implementation and efficiency of the method and the properties of the 159 particles studied here.
- [26] D. Gottwald, G. Kahl, and C. N. Likos, J. Chem. Phys. 122, 204503 (2005).
- [27] S. Gravel and V. Elser, Phys. Rev. E 78, 036706 (2008).
- [28] Y. Jiao and S. Torquato, arXiv:1107.2300v3; P.F. Damasceno, M. Engel, and S.C. Glotzer, arXiv:1109.1323v1.

- [29] B. Wiley, T. Herricks, Y. Sun, and Y. Xia, Nano Lett. 4, 1733 (2004).
- [30] N. Tsapis et al., Phys. Rev. Lett. 94, 018302 (2005).
- [31] K. Brakke, Exp. Math. 1, 141 (1992) [http://www.emis.de/ journals/EM/expmath/volumes/1/1.html].
- [32] M. Gardner, *The Colossal Book of Mathematics: Classic Puzzles, Paradoxes, and Problems* (W. W. Norton & Company, New York, 2001), 1st ed.
- [33] G.F. Tóth, Discrete Comput. Geom. 14, 1 (1995).
- [34] S. Torquato and Y. Jiao, Phys. Rev. E 81, 049908(E) (2010).

Dense Regular Packings of Irregular Nonconvex Particles Supplemental Material

Joost de Graaf,^{1, *} René van Roij,² and Marjolein Dijkstra^{1,†}

¹Soft Condensed Matter, Debye Institute for Nanomaterials Science, Utrecht University, Princetonplein 5, 3584 CC Utrecht, The Netherlands ²Institute for Theoretical Physics, Utrecht University, Leuvenlaan 4, 3584 CE Utrecht, The Netherlands (Dated: September 2, 2011)

Packing Fractions and Crystal Structures for Various Particle Types

In this *Supplemental Material* we present the main body of data we have collected using both our composite technique and literature studies for a large group of solids, particle approximates and several miscellaneous shapes. We also prove that the crystal structures we obtained for rhombicuboctahedra and rhombic enneacontrahedra achieve the densest packing. Furthermore, we present new crystal structures for enneagons, as well as the truncated tetrahedra, which achieve higher packing fractions than previously obtained, both in a centrosymmetric-dimer lattice. In addition to these crystal structures, we consider the relation between the sphericity and packing fraction and show that there is no clear dependence between the two. Finally, we give visual representations for a few of the crystal structures we obtained during our simulation studies.

Method and Systems

Here, we represent the data gathered by our composite technique of the floppy box Monte Carlo method¹ (FBMC), the triangular tessellation method² (TT), and the triangle interference detection method³ (TID). For the TID routine we implemented the *Robust and Accurate Polygon Interference Detection* library³ (RAPID). In our simulations we use a modified criterion to perform lattice reduction⁴

$$(1/18) \cdot (|\mathbf{a}| + |\mathbf{b}| + |\mathbf{c}|) \cdot S(\mathbf{a}, \mathbf{b}, \mathbf{c}) / V(\mathbf{a}, \mathbf{b}, \mathbf{c}) \geq 1.5,$$

with $|\cdot|$ the vector norm; **a**, **b**, and **c** the 3 vectors that span the simulation box; $S(\mathbf{a}, \mathbf{b}, \mathbf{c})$ its surface area; and $V(\mathbf{a}, \mathbf{b}, \mathbf{c})$ its volume.

Systems of tessellated particles were prepared in a dilute phase. By increasing the reduced pressure $p \equiv PV_M/k_BT$ from p = 1 to $p \approx 10^5$ over 50,000 Monte Carlo (MC) cycles we compress the system to a high-density crystalline state. Here P is the pressure, V_M is the volume of a particle model, k_B is the Boltzmann constant, T is the temperature, and one cycle is understood to be one trial move per particle. We typically apply this scheme for each number of particles in the unit cell N (N = 1, ..., 6) and for each considered shape a total of 25 times and select the densest packing among these. These 6 packings (per shape) are allowed to compress for another 10^6 cycles at $p \approx 10^6$, to obtain a maximally compressed state. Finally, we compare these packings and determine the lowest value of N for which the densest packing is achieved and what the lower bound to the packing fraction of the densest packing ϕ_{LB} is, based on our results.

This way of obtaining densest-packed crystal structures is quite efficient. We find excellent agreement with Refs. [5–9] for the Platonic and Archimedean Solids. That is, the system was typically compressed to within 0.002 of the ϕ_{LB} literature value. The simulations we performed yielded a very narrow distribution of crystal-structure candidates near the closest-packed configuration, typically within 1%. Moreover, the method is quite fast. We observed that in the initial 50,000 MC cycles of compression the algorithm exhibits linear scaling. We disregard the final compression run of 10⁶ MC cycles here, since this part only serves to achieve a high decimal accuracy, while the close packed structure no longer changes. Let N_T be the number of triangles of a specific model, $N_C = 50,000$ the number of MC cycles, and T the total run time of the simulation, we obtained

$$\frac{T}{(NN_C)(NN_T)} = \text{constant.}$$

The algorithm thus scales linearly with the total number of triangles times the total number of attempted moves. The value of this constant differs per model, because some models 'crystallize' more easily than others. For the 159 models we studied, we found that the mean value of this constant is ~ 70 μ s with a median value of ~ 40 μ s on a modern 2.0 GHz desktop computer system, with only 27 models exceeding 100 μ s. For more information on the TID algorithm and its benchmarking we refer to Ref. [3].

Tables of Packing Fractions

In Tables I - XI we consider the following quantities: (i) The centrosymmetry of the particle, indicated with 'CS'. 'C' denotes centrosymmetric and 'NC' noncentrosymmetric. (ii) The number of particles N in the unit cell for which densest packing was achieved. (iii) The value of the packing fraction ϕ_{LB} for the densest-known crystal structure. This value has been rounded down to 5 decimals of precision. (iv) The way in which the densest-known packing is accomplished: in a centrosymmetric compound or not. A compound is defined here as an arrangement of particles in space, which are in contact. Our definition is such that the compound may consist of one particle. We abbreviate this parameter by 'CS_c', which assumes the values 'y' for yes, 'n' for no, and '-' for packings where we did not verify this property. (v) Similarly, we determine if the densest-known packing admits a space-filling compound, abbreviated with 'SF_c', which assumes the analogous values 'Y', 'N', and '-'. (vi) The inscribed-sphere upper bound⁷ to the packing fraction $\phi_{\rm UB}$ that we obtained using constrained optimization. (vii, viii) The outscribed-sphere $\phi_{\rm OS}$ and oriented-bounding-box $\phi_{\rm OBB}$ lower bounds to the maximum packing fraction, which were obtained using constrained optimization and the method of Ref. [10]. (ix) The sphericity $\gamma \in [0, 1]$ which we define to be the ratio of the inscribed sphere radius over the outscribed-sphere radius, in analogy to Ref. [7]. We have supplemented the simulation based material with literature results, since most readers will be predominantly interested in the highest $\phi_{\rm LB}$ value. We have put references in the footnotes whenever appropriate - only for 29 out of 159 entries a literature result is known.

TABLE I: Data for the Platonic solids.

Code	\mathbf{CS}	N	$\phi_{ m LB}$	CS_{c}	$\rm SF_{c}$	$\phi_{ m UB}$	$\phi_{ m OS}$	$\phi_{ m OBB}$	γ	name
PS01	NC	4	0.85634^{a}	У	Ν	1.00000	0.09072	0.33333	0.33333	Tetrahedron
PS02	\mathbf{C}	1	0.83635^{b}	У	Ν	0.89343	0.44833	0.51502	0.79465	Icosahedron
PS03	\mathbf{C}	1	0.90450^{b}	У	Ν	0.98116	0.49235	0.47745	0.79465	Dodecahedron
PS04	\mathbf{C}	1	0.94736^{b}	У	\mathbf{Y}^{c}	1.00000	0.23570	0.56218	0.57734	Octahedron
PS05	\mathbf{C}	1	1.00000^{b}	У	\mathbf{Y}^{c}	1.00000	0.27216	1.00000	0.57734	Cube

^aRef. [5]

 b Ref. [6] and [7].

 c Cubes are space filling.^{11,12} Octahedra and tetrahedra form a uniform partition of 3-space (a space-filling compound comprised of Platonic and Archimedean solids) in a 1:2 ratio with equal edge lengths.¹¹

 d The following solids have a nanoparticle or colloid shape equivalent: tetrahedra, $^{13-15}$ cubes, $^{16-18}$ octahedra, 19,20 dodecahedra (macroscopic), 21 and icosahedra. 15,22,23

TABLE II: Data for the Archimedean solids.

Code	\mathbf{CS}	N	$\phi_{ m LB}$	$\mathrm{CS}_{\mathbf{c}}$	SF_{c}	$\phi_{ m UB}$	$\phi_{\rm OS}$	$\phi_{ m OBB}$	γ	name
AS01	NC	2	0.99519^{a}	У	\mathbf{Y}^d	1.00000	0.29718	0.41071	0.52223	Truncated Tetrahedron e
AS02	\mathbf{C}	1	0.78498^{b}	У	Ν	0.83856	0.64230	0.51351	0.91495	Truncated Icosahedron
AS03	NC	1	0.78769^{b}	n^c	Ν	0.93492	0.57484	0.66109	0.85033	$\operatorname{Snub}\operatorname{Cube}^f$
AS04	NC	1	0.78864^{b}	n^c	Ν	0.85547	0.66367	0.53018	0.91886	Snub Dodecahedron
AS05	\mathbf{C}	1	0.80470^{b}	У	Ν	0.83596	0.66075	0.54747	0.92459	Rhombicosidodecahedron
AS06	\mathbf{C}	1	0.82721^{b}	У	Ν	0.89731	0.66498	0.53395	0.90494	Truncated Icosidodecahedron
AS07	\mathbf{C}	1	0.84937^{b}	У	Ν	1.00000	0.59356	0.74491	0.82594	Truncated cuboctahedron
AS08	\mathbf{C}	1	0.86472^{b}	У	Ν	0.93800	0.57737	0.50464	0.85064	Icosidodecahedron
AS09	\mathbf{C}	1	0.87580^{b}	У	Ν	0.87580	0.56262	0.61928	0.86285	${\rm Rhombicuboctahedron}^{g}$
AS10	\mathbf{C}	1	0.89778^{b}	У	Ν	0.97387	0.57413	0.50032	0.83850	Truncated Dodecahedron
AS11	\mathbf{C}	1	0.91836^{b}	У	Ν	1.00000	0.41666	0.83333	0.70710	Cuboctahedron
AS12	\mathbf{C}	1	0.97374^{b}	У	\mathbf{Y}^d	1.00000	0.42712	0.96649	0.67859	Truncated Cube
AS13	\mathbf{C}	1	1.00000^{b}	у	\mathbf{Y}^{d}	1.00000	0.50596	0.53333	0.77459	Truncated Octahedron

 a Ref. [6]

 b Ref. [24] and Ref. [25]. ^cNote that the snub cube and snub dodecahedron are not centrally symmetric, yet they achieve their densest packing in unit cell containing N = 1 particles (Bravais lattice), rather than in a non-Bravais lattice, nor do they form a centrosymmetric compound.

^dTruncated tetrahedra and tetrahedra from a 2:6 space-filling compound with a 3:1 edge length ratio.²⁴ Cuboctahedra and octahedra form a 1:1 uniform partition of 3-space with 1:1 edge length ratio.¹¹ truncated cubes and octahedra form a 1:1 uniform partition of 3-space with edge length ratio 1:1.¹¹

 e For truncated tetrahedra we obtained a new dimer crystal structure, with $\phi_{\rm LB} = 0.98854...$, which was followed by the discovery of the densest packed dimer lattice. 24,25

^fThis result was established using 500 computer experiments for $N = 1, \ldots, 8$ with a slow pressure increase over $4.5 \cdot 10^6$ MC cycles from p = 1 to $p = 1.2^{100}$ in 100 steps, followed by $0.5 \cdot 10^6$ MC cycles of production at that pressure. For all systems we approached the literature value $\phi_{\text{LB}} = 0.78769...$ to within 0.005 and for each N we obtained the same crystal structure, namely the Bravais lattice of Ref. [6], within the numerical uncertainty of our algorithm.

^gWe have demonstrated that Rhombicuboctahedra achieve their densest packing in a crystal lattice: $\phi_{\text{LB}} = (4/3)(4\sqrt{2}-5)$.

^hThe following solids have a nanoparticle or colloid shape equivalent: truncated tetrahedra, 15,26 truncated cubes, 20,26 truncated octahedra, 27 and cuboctahedra. 16,20

	TABLE III. Data for the Catalali solids.													
Code	\mathbf{CS}	N	$\phi_{ m LB}$	$\mathrm{CS}_{\mathbf{c}}$	SF_{c}	$\phi_{ m UB}$	$\phi_{ m OS}$	$\phi_{ m OBB}$	γ	name				
CS01	С	1	0.77155	У	Ν	0.78287	0.61878	0.53980	0.92459	Deltoidal Hexecontahedron				
CS02	\mathbf{C}	1	0.79693	У	Ν	0.85134	0.54691	0.54525	0.86285	Deltoidal Icositetrahedron				
CS03	\mathbf{C}	1	0.79328	У	Ν	0.81365	0.45844	0.54603	0.82594	Disdyakis Dodecahedron				
CS04	\mathbf{C}	1	0.76549	У	Ν	0.77313	0.57295	0.54354	0.90494	Disdyakis Triacontahedron				
CS05	NC	2	0.74107	n^a	Ν	0.78283	0.60732	0.52603	0.91886	Pentagonal Hexecontahedron				
CS06	NC	2	0.74363	n^a	Ν	0.84856	0.52174	0.51407	0.85033	Pentagonal Icositetrahedron				
CS07	\mathbf{C}	1	0.75755	У	Ν	0.78799	0.60356	0.53419	0.91495	Pentakis Dodecahedron				
CS08	\mathbf{C}	1	1.00000	У	\mathbf{Y}^{b}	1.00000	0.35355	0.50000	0.70710	Rhombic Dodecahedron				
CS09	\mathbf{C}	1	0.80174	У	Ν	0.83462	0.51374	0.59016	0.85064	Rhombic Triacontahedron				
CS10	\mathbf{C}	1	0.87601	У	Ν	0.93728	0.29289	0.63158	0.67859	Small Triakis Octahedron				
CS11	\mathbf{C}	1	0.81401	У	Ν	0.87841	0.40824	0.55555	0.77459	Tetrakis Hexahedron				
CS12	\mathbf{C}	1	0.80479	У	Ν	0.81804	0.48227	0.55402	0.83850	Triakis Icosahedron				
CS13	NC	2	0.79886	У	Ν	1.00000	0.16329	0.59999	0.52223	Triakis Tetrahedron				

^aNote that the pentagonal hexecontahedron and pentagonal icositetrahedron are not centrally symmetric, yet these particles do not achieve their densest-known packing by forming a centrosymmetric compound.

^bRhombic dodecahedra are space filling.¹²

^cThe following solids have a nanoparticle or colloid shape equivalent: rhombic dodecahedra^{17,18} and possibly deltoidal icositetrahedra.^{28,29}

 $\ensuremath{\mathrm{TABLE}}\xspace$ IV: Data for the Johnson solids

Code	\mathbf{CS}	N	$\phi_{ m LB}$	CS_{c}	SF_{c}	$\phi_{ m UB}$	$\phi_{\rm OS}$	$\phi_{ m OBB}$	γ	name
JS01	NC	2	0.88745	-	-	1.00000	0.41071	0.49624	0.73848	Augmented Dodecahedron
JS02	NC	2	0.97192	-	-	1.00000	0.21678	0.69255	0.37819	Augmented Hexagonal Prism
JS03	NC	4	0.90463	-	-	1.00000	0.21120	0.66082	0.42422	Augmented Pentagonal Prism
JS04	NC	2	0.83264	-	-	1.00000	0.26330	0.44643	0.57631	Augmented Sphenocorona
JS05	NC	2	0.94527	-	-	1.00000	0.18200	0.57321	0.48671	Augmented Triangular Prism
JS06	NC	2	0.85704	-	-	1.00000	0.13072	0.28916	0.38646	Augmented Tridiminished Icosahedron
JS07	NC	2	0.96347	-	-	1.00000	0.40619	0.85433	0.63827	Augmented Truncated Cube
JS08	NC	1^a	0.87969	-	-	1.00000	0.54646	0.51399	0.81740	Augmented Truncated Dodecahedron
JS09	NC	2	0.90795	-	-	1.00000	0.27695	0.57813	0.57344	Augmented Truncated Tetrahedron
JS10	NC	2	0.90677	-	-	1.00000	0.16543	0.56196	0.37650	Biaugmented Pentagonal Prism
JS11	NC	2	0.91501	-	-	1.00000	0.22322	0.60549	0.48294	Biaugmented Triangular Prism
JS12	С	1	0.96102	У	-	1.00000	0.36374	0.78361	0.59153	Biaugmented Truncated Cube
JS13	NC	2	0.81863	-	-	1.00000	0.62385	0.58749	0.80687	Bigyrate Diminished Rhombicosidodecahedron
JS14	\mathbf{C}	1	0.95273	-	-	1.00000	0.19876	0.62377	0.49112	Bilunabirotunda
JS15	NC	2	0.82232	-	-	1.00000	0.62385	0.57791	0.80687	Diminished Rhombicosidodecahedron
JS16	NC	2	0.85634	-	-	1.00000	0.07654	0.29003	0.33333	Dipyramid 3
JS17	NC	2	0.84024	-	-	1.00000	0.17317	0.32759	0.49112	Dipyramid 5
JS18	NC	2	0.85870	-	-	1.00000	0.37476	0.53256	0.69884	Disphenocingulum
JS19	NC	2	0.83541	-	-	1.00000	0.36461	0.65928	0.45045	Elongated Pentagonal Cupola
JS20	NC	2	0.83751	-	-	1.00000	0.38059	0.46158	0.67091	Elongated Pentagonal Dipyramid
JS21	\mathbf{C}	1	0.79475	у	-	1.00000	0.44920	0.60407	0.60567	Elongated Pentagonal Gyrobicupola
JS22	\mathbf{C}	1	0.81918	у	-	1.00000	0.43524	0.57603	0.74693	Elongated Pentagonal Gyrobirotunda
JS23	NC	2	0.78374	-	-	1.00000	0.51299	0.58594	0.79010	Elongated Pentagonal Gyrocupolarotunda
JS24	NC	2	0.79329	-	-	1.00000	0.44920	0.60407	0.60567	Elongated Pentagonal Orthobicupola
JS25	NC	2	0.81243	-	-	1.00000	0.43524	0.57603	0.74693	Elongated Pentagonal Orthobirotunda
JS26	NC	2	0.79266	-	-	1.00000	0.51299	0.58594	0.79010	Elongated Pentagonal Orthocupolarotunda
JS27	NC	2	0.86656	-	-	1.00000	0.35743	0.53225	0.67555	Elongated Pentagonal Pyramid
JS28	NC	2	0.81652	-	-	1.00000	0.44260	0.61737	0.65993	Elongated Pentagonal Rotunda
JS29	NC	2	0.85746	-	-	1.00000	0.43718	0.68054	0.61012	Elongated Square Cupola
JS30	\mathbf{C}	1	0.90995	у	-	1.00000	0.14788	0.60947	0.41421	Elongated Square Dipyramid
JS31	NC	2	0.80639	-	-	0.87580	0.56262	0.61928	0.86285	Elongated Square Gyrobicupola
JS32	NC	2	0.94371	-	-	1.00000	0.21844	0.72385	0.49999	Elongated Square Pyramid
JS33	NC	2	0.91258	-	-	1.00000	0.35441	0.60017	0.65935	Elongated Triangular Cupola
JS34	NC	2	0.83284	-	-	1.00000	0.05180	0.29326	0.21927	Elongated Triangular Dipyramid
JS35	С	1	0.87941	У	-	1.00000	0.29486	0.62703	0.60243	Elongated Triangular Gyrobicupola
JS36	NC	2	0.88043	-	-	1.00000	0.29486	0.54326	0.60243	Elongated Triangular Orthobicupola
JS37	NC	4	0.86089	-	-	1.00000	0.09737	0.35016	0.28867	Elongated Triangular Pyramid

^aNote that the augmented truncated dodecahedron is not centrally symmetric, yet it achieves its densest-known packing for N = 1 particles in the unit cell.

TABLE V: Data for the Johnson solids - continued.

Code	CS	N	$\phi_{ m LB}$	CS_{c}	SF_{c}	$\phi_{ m UB}$	$\phi_{\rm OS}$	$\phi_{ m OBB}$	γ	name
JS38	NC	2	0.83325	-	-	1.00000	0.58695	0.56431	0.77906	Gyrate Bidiminished Rhombicosidodecahedron
JS39	NC	1^a	0.80470	-	-	0.83596	0.66075	0.54302	0.92459	Gyrate Rhombicosidodecahedron
JS40	NC	2	1.00000	-	\mathbf{Y}^{c}	1.00000	0.15309	0.50000	0.43301	Gyrobifastigium
JS41	NC	2	0.76412	-	-	1.00000	0.42911	0.58293	0.57146	Gyroelongated Pentagonal Bicupola
JS42	NC	2	0.77761	-	-	0.94171	0.45641	0.55737	0.78549	Gyroelongated Pentagonal Birotunda
JS43	NC	4	0.80695	-	-	1.00000	0.34161	0.63982	0.41448	Gyroelongated Pentagonal Cupola
JS44	NC	2	0.78540	-	-	1.00000	0.51719	0.56621	0.78342	Gyroelongated Pentagonal Cupolarotunda
JS45	NC	2	0.86077	-	-	1.00000	0.38637	0.50959	0.64079	Gyroelongated Pentagonal Pyramid
JS46	NC	2	0.81250	-	-	1.00000	0.44203	0.59756	0.63546	Gyroelongated Pentagonal Rotunda
JS47	NC	2	0.77850	-	-	0.97994	0.55378	0.54574	0.82676	Gyroelongated Square Bicupola
JS48	NC	2	0.80712	-	-	1.00000	0.42183	0.60324	0.56972	Gyroelongated Square Cupola
JS49	NC	2	0.80261	-	-	1.00000	0.17614	0.43129	0.51974	Gyroelongated Square Dipyramid
JS50	NC	2	0.82236	-	-	1.00000	0.25752	0.45133	0.59228	Gyroelongated Square Pyramid
JS51	NC	4	0.79162	-	-	1.00000	0.32153	0.52112	0.67198	Gyroelongated Triangular Bicupola
JS52	NC	2	0.83145	-	-	1.00000	0.37306	0.56343	0.64231	Gyroelongated Triangular Cupola
JS53	NC	2	0.83853	-	-	1.00000	0.36444	0.54634	0.62123	Hebesphenomegacorona
JS54	NC	2	0.87796	-	-	1.00000	0.38632	0.51502	0.71464	Metabiaugmented Dodecahedron
JS55	NC	2	0.93602	-	-	1.00000	0.18772	0.65039	0.35100	Metabiaugmented Hexagonal Prism
JS56	NC	2	0.86978	-	-	1.00000	0.53239	0.52766	0.80327	Metabiaugmented Truncated Dodecahedron
JS57	NC	2	0.91942	-	-	1.00000	0.32441	0.46065	0.57232	Metabidiminished Icosahedron
JS58	NC	2	0.83373	-	-	1.00000	0.58695	0.56431	0.77852	Metabidiminished Rhombicosidodecahedron
JS59	NC	1^{b}	0.80470	-	-	0.83596	0.66075	0.54302	0.92459	Metabigyrate Rhombicosidodecahedron
JS60	NC	1^{b}	0.82056	-	-	1.00000	0.62385	0.58749	0.80687	Metagyrate Diminished Rhombicosidodecahedron
JS61	С	1	0.88941	у	-	1.00000	0.33173	0.51502	0.67926	Parabiaugmented Dodecahedron
JS62	С	1	0.97102	у	-	1.00000	0.13937	0.65778	0.31783	Parabiaugmented HexagonalPrism
JS63	С	1	0.88053	у	-	1.00000	0.51540	0.52766	0.79465	Parabiaugmented TruncatedDodecahedron
JS64	С	1	0.85486	у	-	1.00000	0.58695	0.63661	0.68915	Parabidiminished Rhombicosidodecahedron
JS65	С	1	0.80470	у	-	0.83596	0.66075	0.55217	0.92459	Parabigyrate Rhombicosidodecahedron
JS66	NC	1^a	0.82048	-	-	1.00000	0.62385	0.57791	0.80687	Paragyrate Diminished Rhombicosidodecahedron
JS67	NC	2	0.85648	-	-	1.00000	0.09698	0.44385	0.16245	Pentagonal Cupola
JS68	С	1	0.85891	у	-	1.00000	0.19397	0.44385	0.32491	Pentagonal Gyrobicupola
JS69	NC	2	0.84969	-	-	1.00000	0.38567	0.48784	0.58777	Pentagonal Gyrocupolarotunda
JS70	NC	2	0.82381	-	-	1.00000	0.19397	0.44385	0.32491	Pentagonal Orthobicupola
JS71	NC	2	0.81713	-	-	0.93800	0.57737	0.50464	0.85064	Pentagonal Orthobirotunda
JS72	NC	2	0.83123	-	-	1.00000	0.38567	0.48784	0.58777	Pentagonal Orthocupolarotunda
JS73	NC	2	0.85874	-	-	1.00000	0.28868	0.50464	0.42532	Pentagonal Rotunda

 a Note that the gyrate rhombicosidode cahedron and the paragyrate diminished rhombicosidode cahedron are not centrally symmetric, yet they achieve their densest-known packing for N = 1 particles in the unit cell. However, both their densest-known N = 2 packings form a centrosymmetric-dimer lattice, which achieves a packing fraction remarkably close to that of their N = 1 packing.

 b Note that the metabigyrate rhombicosidodecahedron and metagyrate diminished rhombicosidodecahedron are not centrally symmetric, yet the
y achieve their densest-known packing in unit cell containing
 N=1 particles. $^c{\rm The}$ gyrobifastigium is space filling.
 12

TABLE VI: Data for the Johnson solids - continued.

Code	\mathbf{CS}	N	$\phi_{ m LB}$	CS_{c}	SF_{c}	$\phi_{ m UB}$	$\phi_{ m OS}$	$\phi_{ m OBB}$	γ	name
JS74	NC	2	0.94582	-	-	1.00000	0.11785	0.33333	0.36601	Pyramid 4
JS75	NC	2	0.80887	-	-	1.00000	0.08658	0.23032	0.27365	Pyramid 5
JS76	NC	2	0.86477	-	-	1.00000	0.18900	0.65970	0.48676	Snub Disphenoid
JS77	NC	4	0.81981	-	-	1.00000	0.34434	0.52936	0.55150	Snub Square Antiprism
JS78	NC	2	0.82102	-	-	1.00000	0.27733	0.44893	0.58532	Sphenocorona
JS79	NC	2	0.85093	-	-	1.00000	0.16304	0.39771	0.44699	Sphenomegacorona
JS80	NC	2	0.94227	-	-	1.00000	0.15397	0.47140	0.27059	Square Cupola
JS81	NC	2	0.82692	-	-	1.00000	0.30795	0.47140	0.54119	Square Gyrobicupola
JS82	\mathbf{C}	1	0.94249	у	-	1.00000	0.30795	0.55228	0.54119	Square Orthobicupola
JS83	NC	2	0.91836	-	-	1.00000	0.20833	0.41666	0.40824	Triangular Cupola
JS84	NC	2	0.87496	-	-	1.00000	0.26151	0.47213	0.49999	Triangular Hebesphenorotunda
JS85	NC	2	0.88316	-	-	1.00000	0.41666	0.52465	0.70710	Triangular Orthobicupola
JS86	NC	2	0.87421	-	-	1.00000	0.36090	0.52502	0.69033	Triaugmented Dodecahedron
JS87	NC	2	0.89315	-	-	1.00000	0.15008	0.49731	0.31783	Triaugmented Hexagonal Prism
JS88	NC	2	0.82855	-	-	1.00000	0.20411	0.42377	0.50211	Triaugmented Triangular Prism
JS89	NC	2	0.86679	-	-	1.00000	0.52875	0.53355	0.79465	Triaugmented Truncated Dodecahedron
JS90	NC	2	0.91669	-	-	1.00000	0.26245	0.37267	0.50209	Tridiminished Icosahedron
JS91	NC	2	0.84993	-	-	1.00000	0.55005	0.52883	0.73251	Tridiminished Rhombicosidodecahedron
JS92	NC	2	0.80456	-	-	0.83596	0.66075	0.54302	0.92459	Trigyrate Rhombicosidodecahedron

TABLE VII: Data for regular prisms.

Code	CS	N	$\phi_{ m LB}$	CS_{c}	SF_{c}	$\phi_{ m UB}$	$\phi_{ m OS}$	$\phi_{ m OBB}$	γ	name
RP03	NC	2	1.00000^{a}	У	\mathbf{Y}^{c}	1.00000	0.17181	0.50000	0.37796	Prism 3
RP04	С	1	1.00000^{a}	У	\mathbf{Y}^{c}	1.00000	0.27216	1.00000	0.57734	Cube
RP05	NC	2	0.92131^{a}	У	Ν	1.00000	0.31659	0.69098	0.50673	Prism 5
RP06	С	1	1.00000^{a}	У	\mathbf{Y}^{c}	1.00000	0.32863	0.75000	0.44721	Prism 6
RP07	NC	2	0.89269^{a}	У	Ν	1.00000	0.32407	0.73825	0.39803	Prism 7
RP08	С	1	0.90615^{a}	У	\mathbf{Y}^{c}	1.00000	0.31175	0.82842	0.35740	Prism 8
RP09	NC	2	0.90103^{b}	У	Ν	1.00000	0.29629	0.75712	0.32361	Prism 9
RP10	\mathbf{C}	1	0.91371^{a}	У	Ν	1.00000	0.28003	0.77254	0.29524	Prism 10

^aWe used Ref. [30] to compare our results to the literature studies of two-dimensional (2D) regular polygons. See Table I for more information on the cube.

 b For regular enneaprisms (9-gonal base) we have discovered a new densest packing, which also improves upon the result of Ref. [30] for the regular 9-gon (enneagon, nonagon). ^cCubes (square base) and regular tri- (triangular base) and hexaprisms (hexagonal base) are space filling.^{11,12} Octaprisms (8-gonal base)

can form a space-filling compound with irregular triprisms.

TABLE VIII: Data for	regular a	ntiprisms
----------------------	-----------	-----------

Code	\mathbf{CS}	N	$\phi_{ m LB}$	CS_{c}	SF_{c}	$\phi_{ m UB}$	$\phi_{ m OS}$	$\phi_{ m OBB}$	γ	name
AP03	С	1	0.94736	У	Y	1.00000	0.23570	0.56218	0.57734	$Octahedron^a$
AP04	NC	2	0.86343	У	Ν	1.00000	0.30385	0.66666	0.51108	Antiprism 4
AP05	\mathbf{C}	1	0.92052	У	Ν	1.00000	0.32441	0.67418	0.44721	Antiprism 5
AP06	NC	2	0.88189	У	Ν	1.00000	0.32114	0.73204	0.39331	Antiprism 6
AP07	\mathbf{C}	1	0.90137	У	Ν	1.00000	0.30741	0.72740	0.34904	Antiprism 7
AP08	NC	2	0.89332	У	Ν	1.00000	0.28987	0.75526	0.31270	Antiprism 8
AP09	\mathbf{C}	1	0.90672	У	Ν	1.00000	0.27164	0.75000	0.28264	Antiprism 9
AP10	NC	2	0.89731	У	Ν	1.00000	0.25411	0.76608	0.25750	Antiprism 10

 $^a \mathrm{See}$ Table I for more information on the octahedron.

TABLE IX: Data for several miscellaneous solids.

$\begin{array}{c c c c c c c c c c c c c c c c c c c $	
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	е
$MS02 \qquad C \qquad 1 \qquad 1.00000 \qquad y \qquad Y^a \qquad 1.00000 \qquad 0.31426 \qquad 0.66666 \qquad 0.57734 \qquad Elon,$	er's Solid ^{b}
	gated Dodecahedron
MS03 C 1 0.79473 y N 0.79473 0.60457 0.54914 0.91286 Rhor	mbic Enneacontahedron ^c
MS04 C 1 0.82280 y N 1.00000 0.34650 0.52786 0.64945 Rhor	mbic Icosahedron
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$	shed Dodecahedron
MS06 NC 4 0.70503 n N 1.00000 0.13380 0.31616 0.41221 Stan	ford $Bunny^d$
MS07 NC 2 0.47242 y N 1.00000 0.00853 0.06853 0.11355 Ham	merhead \mathbf{Shark}^d

^aThe elongated dodecahedron and the squashed dodecahedron are space filling.

 $^b\mathrm{Note}$ that Dürer's Solid is not the same as the dimer compound formed by truncated tetrahedra.

 c For the rhombic enneacontahedron we have shown that the Bravais lattice we discovered achieves the densest packing.

 d For the Stanford bunny³¹ and the hammerhead shark³² the number of triangles that comprise these models is very high, 3756 and 5116 triangles respectively, however all quantities could be established with the appropriate accuracy.

TABLE X: D	ata for	nonconvex	pol	yhedr	а
------------	---------	-----------	-----	-------	---

Code	\mathbf{CS}	N	$\phi_{ m LB}$	$\mathrm{CS}_{\mathbf{c}}$	SF_{c}	$\phi_{ m UB}$	$\phi_{ m OS}$	$\phi_{ m OBB}$	γ	name
PH01	NC	2	0.61327	n	Ν	1.00000	0.04157	0.23149	0.17469	Császár Polyhedron
PH02	\mathbf{C}	1	0.29477	У	Ν	1.00000	0.07659	0.06269	0.26640	Echidnahedron
PH03	\mathbf{C}	1	1.00000	У	\mathbf{Y}^{a}	1.00000	0.22922	0.45845	0.55284	Escher's Solid
PH04	\mathbf{C}	1	0.55728	У	Ν	1.00000	0.21644	0.20989	0.51160	Great Rhombictriacontrahedron
PH05	\mathbf{C}	2	0.88967	n	Ν	1.00000	0.18806	0.18237	0.18759	Great Stellated Dodecahedron
PH06	\mathbf{C}	1	0.74965	У	Ν	1.00000	0.34558	0.39699	0.53633	Jessen's Orthogonal Icosahedron
PH07	\mathbf{C}	1	0.55602	У	Ν	1.00000	0.20643	0.20019	0.51455	Mathematica Spikey 1^b
PH08	\mathbf{C}	1	0.59998	У	Ν	1.00000	0.14378	0.20246	0.35355	Rhombic Dodecahedron Stellation 2^c
PH09	\mathbf{C}	2	0.55654	n	Ν	1.00000	0.19854	0.19253	0.41946	Rhombic Hexecontrahedron
PH10	\mathbf{C}	2	0.69528	n	Ν	0.97719	0.49635	0.47293	0.79787	Small Triambic Icosahedron
PH11	NC	2	0.51913	У	Ν	1.00000	0.03637	0.13732	0.16538	Szilassi Polyhedron

^{*a*}Escher's solid is space filling by construction.

^bThe number '1' in the name 'Mathematica spikey 1' refers to the first version of the Mathematica spikey, which was used as a logo for the first version of the Mathematica software package.³³ It is a cumulated icosahedron with cumulation ratio $\sqrt{6}/3$.

^cThe number '2' in the name 'rhombic dodecahedron stellation 2' refers to the fact that there are three stellations of the rhombic dodecahedron (four when including the original). This particular stellation is listed as number '2' in the Mathematica polyhedron database.³⁴

TABLE XI: Data for nonconvex nanoparticle and colloid approximates.

Code	\mathbf{CS}	N	$\phi_{ m LB}$	$\mathrm{CS}_{\mathbf{c}}$	SF_{c}	$\phi_{ m UB}$	$\phi_{ m OS}$	$\phi_{ m OBB}$	γ	name
PA01	NC	4	0.51850	n	Ν	1.00000	0.18253	0.27282	0.155754	Cap^b
PA02	\mathbf{C}	1	0.68615	У	Ν	1.00000	0.09602	0.22903	0.38489	Nanostar
PA03	\mathbf{C}	1	0.31077	У	Ν	1.00000	0.02525	0.06681	0.13281	Octapod
PA04	NC	2^a	0.59207	У	Ν	1.00000	0.04864	0.10628	0.20303	Tetrapod

^aThe tetrapod achieves its densest-known packing for N = 2 particles in the unit cell, however, the densest-known N = 1 the packing fraction is remarkably close to that value.

 b The cap³⁵ is comprised of 3850 triangles. Despite this model's complexity, all quantities could be established with the appropriate accuracy.

Densest Packing Configurations for Rhombicuboctahedra and Rhombic Enneacontrahedra

In this section we prove that the densest-packed configurations for rhombicuboctahedra $\phi_{\text{LB}} = (4/3) (4\sqrt{2} - 5)$ and for rhombic enneacontrahedra $\phi_{\text{LB}} = 16 - 34/\sqrt{5}$ are given by their Bravais lattices.

Rhombicuboctahedron: Let the rhombicuboctahedron (RCH) be specified by the vertex coordinates

$$\left(i\left(\frac{1}{2}+\frac{p}{\sqrt{2}}\right), j\left(\frac{1}{2}+\frac{q}{\sqrt{2}}\right), k\left(\frac{1}{2}+\frac{r}{\sqrt{2}}\right)\right),$$

where i, j, and $k \in \{-1, 1\}$ and p, q, and $r \in \{0, 1\}$, with p + q + r = 1. This gives a list of 24 vertices centred on the origin, which span a RCH with volume $4 + 10\sqrt{2}/3$. For this system a possible choice of three vectors which describe a unit cell that realizes the densest packing, is given by

$$v_0 = \left(1 + \frac{1}{\sqrt{2}}, -1 - \frac{1}{\sqrt{2}}, 0\right),$$

$$v_1 = \left(1 + \frac{1}{\sqrt{2}}, 0, -1 - \frac{1}{\sqrt{2}}\right),$$

$$v_2 = \left(0, 1 + \frac{1}{\sqrt{2}}, 1 + \frac{1}{\sqrt{2}}\right).$$

Checking for overlaps in this configuration is a simple matter of verifying that there are no overlaps for an appropriate number of nearest neighbors. It follows that the volume of the unit cell is given by $|v_0 \cdot (v_1 \times v_2)| = 5 + 7/\sqrt{2}$. Therefore, the packing fraction is

$$\phi_{\rm LB} = \frac{4 + 10\sqrt{2}/3}{5 + 7/\sqrt{2}} = \frac{4}{3} \left(4\sqrt{2} - 5 \right)$$

We determined the face-to-point minimum distance for all 26 faces of the RCH, which leads to a set of 26 constrained equations. Using constrained minimization on this set of equations the maximum inscribed sphere can be obtained. Its radius is $1/2 + 1/\sqrt{2}$ and it is centred on the origin. This results in the following upper-bound estimate for the packing fraction

$$\phi_{\rm UB} = \frac{\pi}{\sqrt{18}} \frac{4 + 10\sqrt{2/3}}{\left(4\pi/3\right) \left(1/2 + 1/\sqrt{2}\right)^3} = \frac{4}{3} \left(4\sqrt{2} - 5\right).$$

We have thus proven that the maximum packing fraction is obtained, since $\phi_{\rm UB} = \phi_{\rm LB}$. Here is should be noted that this proof is conditionally dependent on the proof of Ref. [36] via the proof for the upper-bound criterion of Ref. [7].

Rhombic enneacontrahedron: Let the rhombic enneacontrahedron (RECH) be described by the 92 vertex coordinates listed in Tables XII and XIII. These are centred on the origin and span a RECH with volume

$$\frac{20}{3}\sqrt{43+\frac{56\sqrt{5}}{3}}$$

For this system a possible set of three vectors which describes a unit cell that realizes the densest packing, is given by

$$v_{0} = \left(-\frac{5}{6}\left(2+\sqrt{5}\right), \frac{1}{2}\sqrt{\frac{5}{3}}, \frac{1}{3}\left(5+2\sqrt{5}\right)\right),$$

$$v_{1} = \left(-\frac{5}{12}\left(1+\sqrt{5}\right), \sqrt{\frac{235}{24}+\frac{35\sqrt{5}}{8}}, \frac{1}{6}\left(5+\sqrt{5}\right)\right),$$

$$v_{2} = \left(\frac{1}{12}\left(25+13\sqrt{5}\right), \frac{5+\sqrt{5}}{4\sqrt{3}}, \frac{1}{6}\left(5+\sqrt{5}\right)\right).$$

Checking for overlaps in this configuration is again a simple matter. It follows that the volume of the unit cell is given by

$$|v_0 \cdot (v_1 \times v_2)| = \frac{10(20 + 9\sqrt{5})}{3\sqrt{3}}.$$

Therefore, the packing fraction that is achieved for this structure is $\phi_{\text{LB}} = 16 - 34/\sqrt{5}$. By determining the set of 90 face-to-point constrained equations, the maximum inscribed sphere is easily determined to be centred on the origin and have radius

$$\sqrt{\frac{35}{12} + \frac{5\sqrt{5}}{4}}$$

using constrained minimization. This results in the following upper bound to the packing fraction $\phi_{\rm UB} = 16 - 34/\sqrt{5}$. We have thus proven that the maximum packing is obtained.

THE THE THE THOMSTONE CHINCKCONCLUSION
--

$$\begin{array}{ll} \left(\frac{1}{3}\left(-5-\sqrt{5}\right), 0, \frac{1}{3}\left(-7+2\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-7-3\sqrt{5}\right), -\sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{2}{3}\left(-3+\sqrt{5}\right)\right) \\ \left(\frac{1}{6}\left(-7-3\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{2}{3}\left(-3+\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-7-3\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), -1+\frac{2\sqrt{5}}{3}\right) \\ \left(\frac{1}{6}\left(-7-3\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{2}{3}\left(-3+\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-7-3\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), -1+\frac{2\sqrt{5}}{3}\right) \\ \left(\frac{1}{6}\left(-7-3\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{2}{3}\left(-7+\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-7-3\sqrt{5}\right), -\sqrt{\frac{1}{7}}, \frac{4\sqrt{5}}{3}, \frac{1}{3}\left(-7+\sqrt{5}\right)\right) \\ \left(\frac{1}{6}\left(-5-3\sqrt{5}\right), -\sqrt{\frac{7}{6}}, +\frac{\sqrt{5}}{3}, \frac{1}{3}\left(-7+\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-5-3\sqrt{5}\right), -\sqrt{\frac{7}{6}}, +\frac{\sqrt{5}}{3}, \frac{1}{3}\left(-6+\sqrt{5}\right)\right) \\ \left(\frac{1}{6}\left(-5-3\sqrt{5}\right), -\sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), -2\right) & \left(\frac{1}{2}\left(-1-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), -1+\sqrt{5}\right) \\ \left(\frac{1}{6}\left(-7-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), -2\right) & \left(\frac{1}{2}\left(-1-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), -1+\sqrt{5}\right) \\ \left(\frac{1}{6}\left(-7-\sqrt{5}\right), -\sqrt{\frac{1}{6}}, \frac{4\sqrt{5}}{3}, \frac{2}{3}\left(-1+\sqrt{5}\right)\right) & \left(\frac{1}{2}\left(-1-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-7+\sqrt{5}\right)\right) \\ \left(\frac{1}{6}\left(-5-\sqrt{5}\right), -\sqrt{\frac{1}{6}}, \frac{4\sqrt{5}}{3}, \frac{2}{3}\left(-1+\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-7-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-2+\sqrt{5}\right)\right) \\ \left(\frac{1}{6}\left(-5-\sqrt{5}\right), -\sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-2+\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-5-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-7+\sqrt{5}\right)\right) \\ \left(\frac{1}{6}\left(-5-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-2+\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-7-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-7+\sqrt{5}\right)\right) \\ \left(\frac{1}{6}\left(-5-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-2+\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-5-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-7+\sqrt{5}\right)\right) \\ \left(\frac{1}{6}\left(-5-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-7+\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-5-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-7+\sqrt{5}\right)\right) \\ \left(\frac{1}{6}\left(-5-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-2+\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-5-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-7+\sqrt{5}\right)\right) \\ \left(\frac{1}{6}\left(-5-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-7+\sqrt{5}\right)\right) & \left(\frac{1}{6}\left(-5-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-7+\sqrt{5}\right)\right) \\ \left(\frac{1}{6}\left(-5-\sqrt{5}\right), \sqrt{\frac{1}{6}}\left(3+\sqrt{5}\right), \frac{1}{3}\left(-7+\sqrt{5}\right)\right) & \left(\frac{1}{$$

$$\begin{array}{c} \left(-\frac{1}{3}, \sqrt{3} + \frac{4\sqrt{5}}{3}, \frac{1}{3} \left(-3 + \sqrt{5} \right) \right) \\ \left(\frac{1}{6} \left(-3 + \sqrt{5} \right), \sqrt{\frac{1}{6} \left(3 + \sqrt{5} \right)}, -\frac{1}{3} + \sqrt{5} \right) \\ \left(0, -\sqrt{\frac{2}{3} \left(3 + \sqrt{5} \right)}, -1 + \sqrt{5} \right) \\ \left(0, -\sqrt{\frac{2}{3} \left(3 + \sqrt{5} \right)}, -1 + \sqrt{5} \right) \\ \left(\frac{1}{6} \left(3 - \sqrt{5} \right), \sqrt{\frac{1}{6} \left(3 + \sqrt{5} \right)}, -\frac{8}{3} \right) \\ \left(\frac{1}{3}, -\sqrt{3} + \frac{4\sqrt{5}}{3}, -1 + \frac{2\sqrt{5}}{3} \right) \\ \left(\frac{1}{3}, \sqrt{3} + \frac{4\sqrt{5}}{3}, -1 + \frac{2\sqrt{5}}{3} \right) \\ \left(\frac{1}{6} \left(1 + \sqrt{5} \right), \sqrt{\frac{5}{6} \left(3 + \sqrt{5} \right)}, \frac{1}{3} \left(-7 + \sqrt{5} \right) \right) \\ \left(\frac{\sqrt{5}}{3}, -\frac{1}{\sqrt{3}}, -\frac{8}{3} \right) \\ \left(\frac{\sqrt{5}}{3}, \sqrt{\frac{5}{3}}, -\frac{2}{3} + \sqrt{5} \right) \\ \left(\frac{1}{6} \left(3 + \sqrt{5} \right), -\sqrt{\frac{1}{6} \left(3 - \sqrt{5} \right)}, -\frac{1}{3} + \sqrt{5} \right) \\ \left(\frac{1}{6} \left(3 + \sqrt{5} \right), \sqrt{\frac{2}{3} \left(3 + \sqrt{5} \right)}, \frac{2}{3} \left(-1 + \sqrt{5} \right) \right) \\ \left(\frac{1}{6} \left(5 + \sqrt{5} \right), \sqrt{\frac{5}{6} \left(3 + \sqrt{5} \right)}, \frac{2}{3} \left(-2 + \sqrt{5} \right) \right) \\ \left(\frac{1}{6} \left(5 + \sqrt{5} \right), -\sqrt{\frac{7}{6} \left(5 + \sqrt{5} \right)}, \frac{\sqrt{5}}{6} \left(3 + \sqrt{5} \right)}, \frac{2}{3} \left(-2 + \sqrt{5} \right) \right) \\ \left(\frac{1}{6} \left(7 + \sqrt{5} \right), -\sqrt{\frac{7}{6} \left(5 + \sqrt{5} \right)}, \frac{2}{3} \left(-2 + \sqrt{5} \right) \right) \\ \left(\frac{1}{6} \left(7 + \sqrt{5} \right), -\sqrt{\frac{7}{6} \left(5 + \sqrt{5} \right)}, \frac{2}{3} \left(-2 + \sqrt{5} \right) \right) \\ \left(\frac{1}{6} \left(7 + \sqrt{5} \right), -\sqrt{\frac{7}{6} \left(5 + \sqrt{5} \right)}, -2 \right) \\ \left(\frac{1}{3} \left(3 + \sqrt{5} \right), 0, -\frac{2}{3} + \sqrt{5} \right) \\ \left(\frac{1}{6} \left(5 + 3\sqrt{5} \right), -\sqrt{\frac{7}{6} \left(5 + \sqrt{5} \right)}, -1 + \frac{2\sqrt{5}}{3} \right) \\ \left(\frac{1}{6} \left(5 + 3\sqrt{5} \right), -\sqrt{\frac{7}{6} \left(3 + \sqrt{5} \right)}, -1 + \frac{2\sqrt{5}}{3} \right) \\ \left(\frac{1}{6} \left(5 + 3\sqrt{5} \right), -\sqrt{\frac{7}{6} \left(3 + \sqrt{5} \right)}, \frac{1}{3} \left(-6 + \sqrt{5} \right) \right) \\ \left(\frac{1}{6} \left(7 + 3\sqrt{5} \right), -\sqrt{\frac{1}{6} \left(3 + \sqrt{5} \right)}, \frac{1}{3} \left(-6 + \sqrt{5} \right) \right) \\ \left(\frac{1}{6} \left(7 + 3\sqrt{5} \right), \sqrt{\frac{1}{6} \left(3 + \sqrt{5} \right)}, \frac{1}{3} \left(-6 + \sqrt{5} \right) \right) \\ \left(\frac{1}{3} \left(5 + \sqrt{5} \right), 0, \frac{1}{3} \left(-5 + \sqrt{5} \right) \right) \end{array}$$

$$\frac{1}{6} (-3 + \sqrt{5}), -\sqrt{\frac{1}{6}} (3 + \sqrt{5}), -\frac{1}{3} + \sqrt{5}) \\ (0, -\sqrt{\frac{2}{3}} (3 + \sqrt{5}), -2) \\ (0, 0, -3) \\ (0, 0, -3) \\ (0, \sqrt{\frac{2}{3}} (3 + \sqrt{5}), -2) \\ (\frac{1}{6} (3 - \sqrt{5}), -\sqrt{\frac{1}{6}} (3 + \sqrt{5}), -\frac{8}{3}) \\ (\frac{1}{3}, -\sqrt{3 + \frac{4\sqrt{5}}{3}}, \frac{2}{3}} (-3 + \sqrt{5})) \\ (\frac{1}{3}, \sqrt{3 + \frac{4\sqrt{5}}{3}}, \frac{2}{3}} (-3 + \sqrt{5})) \\ (\frac{1}{4}, \sqrt{3 + \frac{4\sqrt{5}}{3}}, \frac{2}{3}} (-3 + \sqrt{5})) \\ (\frac{1}{6} (1 + \sqrt{5}), -\sqrt{\frac{5}{6}} (3 + \sqrt{5}), \frac{1}{3} (-7 + \sqrt{5})) \\ (\frac{\sqrt{5}}{3}, -\sqrt{\frac{5}{3}}, -\frac{2}{3} + \sqrt{5}) \\ (\frac{\sqrt{5}}{3}, \frac{1}{\sqrt{3}}, -\frac{8}{3}) \\ (\frac{1}{6} (3 + \sqrt{5}), -\sqrt{\frac{7}{6}} + \frac{\sqrt{5}}{2}, -\frac{7}{3}) \\ (\frac{1}{6} (3 + \sqrt{5}), \sqrt{\frac{1}{6}} (3 - \sqrt{5}), -\frac{1}{3} + \sqrt{5}) \\ (\frac{1}{3} (1 + \sqrt{5}), -\sqrt{\frac{2}{3}} (3 + \sqrt{5}), \frac{2}{3} (-1 + \sqrt{5})) \\ (\frac{1}{6} (5 + \sqrt{5}), -\sqrt{\frac{5}{6}} (3 + \sqrt{5}), \frac{1}{3} (-7 + 2\sqrt{5})) \\ (\frac{1}{6} (5 + \sqrt{5}), \sqrt{\frac{5}{6}} (3 + \sqrt{5}), \frac{1}{3} (-7 + 2\sqrt{5})) \\ (\frac{1}{2} (1 + \sqrt{5}), -\sqrt{\frac{1}{6}} (3 + \sqrt{5}), -1 + \sqrt{5}) \\ (\frac{1}{2} (1 + \sqrt{5}), -\sqrt{\frac{1}{6}} (3 + \sqrt{5}), -1 + \sqrt{5}) \\ (\frac{1}{2} (1 + \sqrt{5}), -\sqrt{\frac{7}{6}} + \frac{\sqrt{5}}{2}, \frac{2}{3} (-3 + \sqrt{5})) \\ (\frac{1}{6} (5 + 3\sqrt{5}), -\sqrt{\frac{7}{6}} + \frac{\sqrt{5}}{2}, \frac{2}{3} (-3 + \sqrt{5})) \\ (\frac{1}{6} (7 + 3\sqrt{5}), -\sqrt{\frac{1}{6}} (3 + \sqrt{5}), \frac{1}{3} (-7 + \sqrt{5})) \\ (\frac{1}{6} (7 + 3\sqrt{5}), -\sqrt{\frac{1}{6}} (3 + \sqrt{5}), \frac{1}{3} (-3 + \sqrt{5})) \\ (\frac{1}{6} (7 + 3\sqrt{5}), -\sqrt{\frac{1}{6}} (3 + \sqrt{5}), \frac{1}{3} (-3 + \sqrt{5})) \\ (\frac{1}{6} (7 + 3\sqrt{5}), -\sqrt{\frac{1}{6}} (3 + \sqrt{5}), \frac{1}{3} (-3 + \sqrt{5})) \\ (\frac{1}{6} (7 + 3\sqrt{5}), -\sqrt{\frac{1}{6}} (3 + \sqrt{5}), \frac{1}{3} (-3 + \sqrt{5})) \\ (\frac{1}{6} (7 + 3\sqrt{5}), \sqrt{\frac{1}{6}} (3 + \sqrt{5}), \frac{1}{3} (-3 + \sqrt{5})) \\ (\frac{1}{6} (7 + 3\sqrt{5}), \sqrt{\frac{1}{6}} (3 + \sqrt{5}), \frac{1}{3} (-3 + \sqrt{5})) \\ (\frac{1}{3} (5 + \sqrt{5}), 0, \frac{1}{3} (-2 + \sqrt{5})) \end{cases}$$

New Crystal Structures for Enneagons and Truncated Tetrahedra

In this section we describe the construction of a new crystal structure for enneagons (nonagons, regular 9-gon), which achieves the densest-known packing. The method we use here has similarities the ones employed in Refs. [5] and [9]; both of these present the analytic construction of a dense family of tetrahedral dimers. Finally, we list our numerical data, by which the dimer lattice of truncated tetrahedra can be constructed.

Enneagons: We begin with some basic definitions. An enneagon is defined here to have a centre-to-tip distance of 1. It is centred on the origin of a Cartesian coordinate system with its tips mirror-symmetrically distributed around the y-axis and one tip located on the positive y-axis. To describe the crystal structure we further require three two-dimensional (2D) vector parameterizations

$$p_{1}(q) = \left(q\left(\sin\left[\frac{\pi}{9}\right] - \frac{\sqrt{3}}{2}\right) + 2\cos\left[\frac{\pi}{9}\right]\sin\left[\frac{2\pi}{9}\right], \frac{1}{4}\left(q\left(2 - 4\cos\left[\frac{\pi}{9}\right]\right) - \csc\left[\frac{\pi}{18}\right]\right)\right),$$

$$p_{2}(r) = \left(2\cos\left[\frac{\pi}{9}\right]\sin\left[\frac{\pi}{9}\right] - r\sin\left[\frac{2\pi}{9}\right], 2\cos^{2}\left[\frac{\pi}{9}\right] + r\left(1 - \cos\left[\frac{2\pi}{9}\right]\right)\right),$$

$$p_{3}(s) = \left(s\left(\sin\left[\frac{\pi}{9}\right] - \frac{\sqrt{3}}{2}\right) + 2\cos\left[\frac{\pi}{9}\right]\sin\left[\frac{2\pi}{9}\right], -\frac{1}{4}\left(s\left(2 - 4\cos\left[\frac{\pi}{9}\right]\right) - \csc\left[\frac{\pi}{18}\right]\right)\right)$$

with $q, r, and s \in [-1, 1]$. We will employ these to describe lattice vectors and positions of the enneagons in the unit cell. We eliminate two of the variables such that different enneagons in the lattice have some of their edges and corners touch and slide over each other upon varying the third:

$$\begin{split} T(k,l) &= \frac{\csc\left[\frac{2\pi}{9}\right]}{4\left(\cos\left[\frac{2\pi}{9}\right] - 1\right)} \cdot \\ &\left\{\sqrt{3}\left(\cos\left[\frac{2\pi}{9}\right] - 1\right)(k+l) + 2\sin\left[\frac{\pi}{9}\right](k+l) - 8\sin\left[\frac{\pi}{9}\right]\cos\left[\frac{\pi}{9}\right] \\ &- \sin\left[\frac{\pi}{9}\right]\cos\left[\frac{2\pi}{9}\right](k+l) + 8\sin\left[\frac{\pi}{9}\right]\cos\left[\frac{\pi}{9}\right]\cos\left[\frac{2\pi}{9}\right] + \sin\left[\frac{2\pi}{9}\right](l-k) \\ &- 2\sin\left[\frac{2\pi}{9}\right]\cos\left[\frac{\pi}{9}\right](l-k) + 8\sin\left[\frac{2\pi}{9}\right]\cos\left[\frac{\pi}{9}\right] - 8\sin\left[\frac{2\pi}{9}\right]\cos\left[\frac{\pi}{9}\right]\cos\left[\frac{\pi}{9}\right] \\ U(k) &= \left\{\sqrt{3}k + 2\cos\left[\frac{\pi}{18}\right] - k\cos\left[\frac{\pi}{18}\right] - \sqrt{3}k\cos\left[\frac{\pi}{9}\right] + 2k\cos\left[\frac{\pi}{18}\right]\cos\left[\frac{\pi}{9}\right] + \sqrt{3}k\sin\left[\frac{\pi}{18}\right] - k\sin\left[\frac{\pi}{9}\right] \\ &4\sin\left[\frac{\pi}{18}\right]\cos\left[\frac{\pi}{18}\right] - 2k\sin\left[\frac{\pi}{18}\right]\sin\left[\frac{\pi}{9}\right] - 4\sin\left[\frac{2\pi}{9}\right]\cos\left[\frac{\pi}{9}\right] - 8\sin\left[\frac{\pi}{18}\right]\sin\left[\frac{2\pi}{9}\right]\cos\left[\frac{\pi}{9}\right] \right\} \\ V(k) &= \sin\left[\frac{\pi}{19}\right] + 2\sin\left[\frac{\pi}{18}\right]\sin\left[\frac{\pi}{9}\right] - \cos\left[\frac{\pi}{18}\right] - \sqrt{3}\left(\sin\left[\frac{\pi}{18}\right] + \cos\left[\frac{\pi}{9}\right]\right) + 2\cos\left[\frac{\pi}{18}\right]\cos\left[\frac{\pi}{9}\right] \right\} \\ W(k) &= \frac{U(k)}{V(k)}, \end{split}$$

with k and $l \in [-1, 1]$. Using T(k, l) and W(k), we may write

$$P_{0}(k) = (0,0),$$

$$P_{1}(k) = p_{2} (T (k, W (k))),$$

$$V_{0}(k) = p_{3} (W (k)) + p_{2} (T (k, W (k))),$$

$$V_{1}(k) = p_{1} (k) + p_{3} (W (k)),$$

where the P_i give the position of the enneagons in the unit cell (N = 2) with lattice vectors V_i $(i \in \{0, 1\})$. The enneagon at P_0 has the same orientation as the base enneagon defined above and the one at P_1 is rotated by π with respect to the base enneagon, also see Fig. 1a which shows this configuration for the densest-known packing. By determining the value of k, say k^* , for which the volume fraction F_v associated to this lattice is maximized,

$$F_{v}(k) = \frac{18 \sin\left\lfloor\frac{\pi}{9}\right\rfloor \cos\left\lfloor\frac{\pi}{9}\right\rfloor}{|V_{0,x}(k)V_{1,y}(k) - V_{0,y}(k)V_{1,x}(k)|},$$

we obtain the lattice with the highest-known packing fraction. For this value of k, we obtain the following

$$\begin{aligned} k^* &= 0.334782056761309\dots, \\ F_v(k^*) &= 0.901030078420934\dots = \phi_{\text{LB}}, \\ P_0(k^*) &= (0,0), \\ P_1(k^*) &= (0.8471436672437109\dots, 1.691664920976177\dots), \\ V_0(k^*) &= (1.7675368645589482\dots, 3.372726522382239\dots), \\ V_1(k^*) &= (1.9530111855752121\dots, 0.094167780690677\dots). \end{aligned}$$

This results in the following 2D crystal structure, see Fig. 1, which shows the unit cell and a piece of the crystal this generates. Note that we have confirmed that at least one the packings of Ref. [30] can be impoved upon by large scale reorganizations. Also note that this configuration forms a centrosymmetric-dimer lattice.



FIG. 1: New densest-packing crystal structure for enneagons. (a) The unit cell vectors $V_0(k^*)$ and $V_1(k^*)$ and the N = 2 enneagons in it, positioned at $P_0(k^*)$ and $P_1(k^*)$. (b) A piece of the crystal structure this dimer generates.

Truncated tetrahedra: For the system containing N = 2 truncated tetrahedra we provide additional information on the composition of the dimer lattice that we obtained using our method and which achieves a packing fraction $\phi_{\text{LB}} = 0.988...$ Table XIV lists the position and orientation of the particles within the unit cell, as well as the shape of the cell itself. We have transformed the unit cell of the dimer lattice (N = 2), using lattice reduction⁴, in such a way that it is almost cubic and that one of the particles is located in the origin.

TABLE XIV: Coordinates which specify the dimer lattice of truncated tetrahedra. This Table lists the 12 vertices \mathbf{v} of the truncated tetrahedron model used in our simulations. It also gives the three vectors \mathbf{u}_m , with m = 1, 2, 3 an index, which span the unit cell; the two position vectors \mathbf{R}_i , with i = 1, 2 the particle number, which indicate where the truncated tetrahedra are located with respect to the origin; and the two rotation matrices \mathbf{M}_i , which specify how to rotate the particles from their initial configuration. This initial configuration is given by the set of \mathbf{v} presented here. A single vertex is a three dimensional (3D) vector, of which the components are indicated by v_x, v_y , and v_z , relative to a standard Cartesian coordinate frame. These \mathbf{v} have been written in a row format in the Table, other vectors are treated similarly. The entries of the matrices \mathbf{M} are denoted as M_{kl} , with k, l = x, y, z. For this choice of vertices, the volume enclosed by the particle's surface is unity. We have provided all vector and matrix entries in 6 decimal precision. Rounding errors may lead to small overlaps of particles in the crystal generated using these coordinates.

v_x		v_y	v_z		v_x	v_y		v_z		u_x, R_x	u_y, R_y	u_z, R_z
0.6211	21 -0.3	358604	-0.4392	00 0	.621121	0.358	3604 -0	-0.439200	\mathbf{u}_1	0.241977	0.928872	0.855892
0.8281	62 0.0	00000	0.1464	00 -0	.414081	-0.717	209	0.146400	\mathbf{u}_2	0.604353	-0.735843	0.832841
-0.6211	.21 -0.3	358604	-0.4392	00 0	.000000	-0.717	209 -	-0.439200	\mathbf{u}_3	-1.053988	-0.200499	0.654313
0.0000	00 0.7	17209	-0.4392	-00	.621121	0.3580	- 304 -	-0.439200				
-0.4140	0.7 0.7	17209	0.1464	-00	.207040	0.3580	304	0.732000	\mathbf{R}_1	0.000000	0.000000	0.000000
-0.2070	-0.3	358604	0.7320	0 0	.414081	0.000	000	0.732000	\mathbf{R}_2	-0.073508	-0.001753	0.875316
	M_{xx}	Ι	M_{xy}			M_{yx}	M_{yy}	i	M_{yz}	M_{zx}	M_{zy}	M_{zz}
\mathbf{M}_1	$M_1 -0.892816$		-0.442579 0.		-0.443985		0.8960	32 0.0	01996	-0.075867	-0.035373	-0.996490

0.896032

-0.001996

0.075867

-0.035373

0.996490

0.443985

 \mathbf{M}_2

0.892816

-0.442579

-0.083685

Visual Representations of the Closest Packing Results

In this section we visually represent the data of Tables I - XI to show that there is no clear relation between the sphericity γ and the densest packing fraction ϕ_{LB} for the 159 particle species that we have investigated. Here, we also show that for the convex particles Ulam's conjecture is satisfied. Finally, we give more extensive visual representations of some of the data in Tables III, IX and X by showing several of the packings achieved using our method.



FIG. 2: Packing fraction for the densest-known configuration of a particle and the relation to its sphericity. The graph shows the achieved maximum packing fraction ϕ_{LB} as a function of the sphericity γ for the convex particles (circles, blue) and nonconvex particles (crosses, red) we investigated. Also see Tables I - XI for the numerical value associated with this data. Note that particles with a sphericity of $\gamma > 0.8$ tend to group closer to the packing fraction of spheres (ϕ_{SPH} , solid line). However, there is significant spread in the ϕ_{LB} for all particles we considered, even for $\gamma > 0.8$. Therefore, we conclude that there is no clear relation between γ and ϕ_{LB} on the strength of our data. Using the line ϕ_{SPH} and the inset, we show that all convex particles satisfy Ulam's conjecture.



FIG. 3: Upper and lower bounds to the densest packing fraction. Graph (a) shows bounds to the densest packing fraction for 13 Catalan solids and graph (b) shows analogous data for 13 nonconvex solids. The graphs show the achieved densest-known packing fraction (ϕ_{LB} , connected crosses), as well as the outscribed-sphere lower bound (ϕ_{OS} , circles), oriented-bounding-box lower bound (ϕ_{OBB} , squares), and inscribed-sphere upper bound⁷ (ϕ_{UB} , diamonds) values to the packing fraction for the models given below it. Also see Table III, IX and X, which gives both the numerical values and the full name corresponding to the abbreviations used here. Based on the available data we expect to find our FBMC result inside the gray area, which is bounded from below and above by the established lower and upper bound to the densest packing fraction respectively. The value of the densest packing for spheres ϕ_{SPH} is indicated by a red line. Note that the improvement of the FBMC method with respect to the established lower bounds is significant for all models.



FIG. 4: Several crystal structures achieved for a model of a colloidal cap. This figure shows two views of the cap model used in our simulations (a) - (b), which is representative of colloidal caps obtained in syntheses, see for instance Refs. [37] and [38]. The cap model is derived from the numerical analysis of the collapse of a spherical shell. This analysis^{35,39} was performed using Surface Evolver⁴⁰ to minimize the Hamiltonian, which describes the properties of the shell. The Hamiltonian incorporates bending and in-plane stretching elasticity terms to properly account for the physics behind the collapse under an external isotropic pressure. Note the buckling that has occurred in the impression left by the shell collapse (b). Also note that the model is not rotationally symmetric (b). We find several crystal-structure candidates. For N = 1 we find a columnar phase (c); the 26 of its periodic images are shown. For N = 2, 3, 4, and 5, we obtain braided phases without inversion, such phases are labeled 'B' in Ref. [41]. The unit cell and crystal structure for N = 4 particles in the unit cell are shown in (d) and (e) respectively, where we have labeled the different caps with colors. The structure is a binary braided configuration; only 7 periodic images are shown. The binary nature is likely due to the lack of rotational symmetry, which allows for better packing. Finally, for N = 6 we obtain a rough braided phase with inversions (f), which looks similar to the 'IB phase' predicted in Ref. [41]; again only 7 periodic images are shown and colors were used to aid in indentifying the periodicity. Because of the substantial difference in shape to the bowl-shaped particles used in Ref. [41], we do not think it appropriate to assign an approximate L/σ (see Ref. [41]) value to the cap model.



FIG. 5: The densest known regular packing of hammerhead shark models. Different views (a) - (e) of hammerhead shark model.³² The unit cell of the densest regular packing ($\phi_{\text{LB}} = 0.472$) is shown in (f), and a piece of the crystal in (g). The crystal structure is a double lattice where two hammerhead sharks (red, blue) point in opposite directions and one is rotated by an angle of ~ π radians around its long axis with respect to the other, thereby forming a centrosymmetric dimer.

- * Electronic address: j.degraaf1@uu.nl
- [†] Electronic address: m.dijkstra1@uu.nl
- ¹ L. Filion et al., Phys. Rev. Lett. **103**, 188302 (2009).
- $\mathbf{2}$ J. de Graaf, M. Dijkstra, and R. van Roij, Phys. Rev. E 80, 051405 (2009).
- ³ GAMMA Research Group at the University of North Carolina, RAPID Robust and Accurate Polygon Interference Detection, http://gamma.cs.unc.edu/OBB/, 1997.
- ⁴ D. Gottwald, G. Kahl, and C. N. Likos, J. Chem. Phys. **122**, 204503 (2005).
- ⁵ E. R. Chen, M. Engel, and S. C. Glotzer, Discrete Comput. Geom. 44, 253 (2010).
- ⁶ U. Betke and M. Henk, Comput. Geom. **16**, 157 (2000).
- ⁷ S. Torquato and Y. Jiao, Nature Lett. **460**, 876 (2009).
- ⁸ S. Torquato and Y. Jiao, Phys. Rev. E **80**, 041104 (2009).
- ⁹ S. Torquato and Y. Jiao, Phys. Rev. E **81**, 041310 (2010).
- ¹⁰ S. Gottschalk, M. C. Lin, and D. Manocha, OBBTree: a hierarchical structure for rapid interference detection, in *Proceedings* of the 23rd annual conference on Computer graphics and interactive techniques, SIGGRAPH '96, page 171, Association for Computing Machinery (New York), 1996.
- ¹¹ G. Vernizzi, R. Sknepnek, and M. Olvera de la Cruz, Europ. J. Combinatorics **21**, 807 (2000).
- ¹² U. Agarwal and F. A. Escobedo, Nat. Mater. **10**, 230 (2011).
- ¹³ A. Mews, A. V. Kadavanich, U. Banin, and A. P. Alivisatos, Phys. Rev. B 53, R13242 (1996).
- ¹⁴ E. C. Greyson, J. E. Barton, and T. W. Odom, Small **2**, 368 (2006).
- ¹⁵ F. Kim et al., Angew. Chem. **116**, 3759 (2004).
- ¹⁶ Y. Sun and Y. Xia, Science **298**, 2176 (2002).
- ¹⁷ X. Zhang et al., Angew. Chem. **48**, 9121 (2009).
- ¹⁸ H.-L. Wu, C.-H. Kuo, and M. H. Huang, Langmuir **26**, 12307 (2010).
- ¹⁹ E. Matijevic, Acc. Chem. Res. **14**, 22 (1981).
- ²⁰ A. Tao, P. Sinsermsuksakul, and P. Yang, Angew. Chem. 45, 4597 (2006).
- ²¹ I. R. Fisher et al., Phys. Rev. B **59**, 308 (1999).
- ²² K. Kwon et al., J. Phys. Chem. C **111**, 1161 (2007).
- ²³ J. Tang et al., J. Am. Chem. Soc. **129**, 9044 (2007).
 ²⁴ Y. Jiao and S. Torquato, Arxiv **1107.2300v3**, na (2011).
- ²⁵ P. F. Damasceno, M. Engel, and S. C. Glotzer, Arxiv **1109.1323v1**, na (2011).
- ²⁶ B. Wiley, T. Herricks, Y. Sun, and Y. Xia, Nano Lett. 4, 1733 (2004). 27
- A. S. Barnard, X. M. Lin, and L. A. Curtiss, J. Phys. Chem. B 109, 24465 (2005).
- ²⁸ Y. Saito, K. Mihama, and R. Uyeda, J. Cryst. Growth **45**, 501 (1978).
- ²⁹ Y. Saito, K. Mihama, and R. Uyeda, Jpn. J. Appl. Phys. **19**, 1603 (1980).
- ³⁰ Y. Limon Duparcmeur, A. Gervois, and J. P. Troadec, J. Phys. I France 5, 1539 (1995).
- ³¹ Stanford University Computer Graphics Laboratory, Stanford Bunny PLY-model, http://graphics.stanford.edu/data/ 3Dscanrep/, 1993.
- ³² J. Burkardt, Hammerhead Shark PLY-model, http://people.sc.fsu.edu/~jburkardt/data/ply/ply.html, 1993.
- ³³ Wolfram Research, Inc., Mathematica 1, Software Package, 1988.
- ³⁴ Wolfram Research, Inc., Mathematica 7, Software Package, 2009.
- ³⁵ C. Quilliet et al., Eur. Phys. J. E **27**, 13 (2008).
- ³⁶ T. C. Hales and S. P. Ferguson, Discete Comput. Geom. **36**, 1 (2006).
- ³⁷ C. I. Zoldesi and A. Imhof, Adv. Mater. **17**, 924 (2005).
- ³⁸ D. Jagadeesan et al., Angew. Chem. **47**, 7685 (2008).
- ³⁹ N. Tsapis et al., Phys. Rev. Lett. **94**, 018302 (2005).
- ⁴⁰ K. Brakke, Exp. Math. **1**, 141 (1992).
- ⁴¹ M. Marechal et al., Nano Lett. **10**, 1907 (2010).