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Random Loose Packing in Granular Matter

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Abstract We introduce and simulate a two-dimensional Edwards-style model of granular matter at vanishing pressure. The model incorporates some of the effects of gravity and friction, and exhibits a random loose packing density whose standard deviation vanishes with increasing system size, a phenomenon that should be verifiable for real granular matter.

Keywords Granular matter · Random loose packing

1 An Edwards-Style Model

We introduce and analyze a crude model for the random loose packings of granular matter. These packings, as well as random close packings, were carefully prepared by Scott et al. in the 1960s [17, 18], in samples of steel ball bearings. Gently pouring samples of 20,000 to 80,000 spheres into a container, the lowest possible volume fraction obtainable—the so-called random loose packing density—was determined to be 0.608 ± 0.006.

The above refers to monodisperse steel spheres immersed in air; they also worked with spheres of other materials immersed in other fluids; variations in the coefficient of friction and in the effective gravitational force lead to somewhat different values for the random loose packing density [18].

Matter is generally described as "granular" if it is composed of a large number of non-cohesive subunits each of which is sufficiently massive that its gravitational energy is much larger than its thermal energy. A common example is a sand pile.

There are several classic phenomena characteristic of static granular matter, in particular dilatancy, random close packing, and random loose packing, none of which can yet be considered well-understood; see [3] for a good review. A basic question about these phenomena

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is whether they are sharply defined or inherently vague. Dilatancy has recently been associated with a phase transition measured by the response of the material to shear [19], which answers the question for this phenomenon. The case of random close packing is controversial and awaits further experiment; see [16, 21]. Our main goal here is to analyze this question with respect to random loose packing, to determine whether or not traditional theoretical approaches to granular matter predict a sharply defined random loose packing density. It is clear that any experimental determination of a random loose packing density will vary with physical conditions such as coefficient of friction, and we will take this variation into account in our analysis below.

We begin by contrasting two common approaches to modelling static granular matter. One, the more common, is the "protocol-dependent simulation," in which one studies properties of dense packings by exploring a variety of methods of preparation of the packings; see [5, 21] or [23] for examples. Another approach goes under the name of Edwards theory [4], in which, basically, one adds the effects of friction and a strong gravitational force to the hard sphere model of equilibrium statistical mechanics. We note that appropriate specification of the added forces fully determines an Edwards model; there are no adjustable parameters beyond those familiar from statistical mechanics, such as density and pressure. (Of course one can always introduce further approximations or features, for instance mean field theory, soft core, attraction, etc.) In particular, in an Edwards model all Markov chain Monte Carlo simulations will, if done correctly, give the same result; there is no freedom in preparing the packings the way there is in protocol-dependent simulation.

These two approaches—the protocol-dependent simulations and the Edwards approach—have different strengths. There have been serious claims that the former approach has serious difficulty making sense of some granular phenomena, in particular random close packing [21]. We have previously shown [16] how Edwards theory allows a clean definition of random close packing, and in this paper we show, by a very different mechanism, how it allows for an understanding of random loose packing. In that sense our choice of using an Edwards-type model is central to our argument. (We do not claim that the Edwards approach has been proven the most accurate theory of static granular matter, but only that it is a serious contender.)

We briefly summarize our Edwards-style model as follows: We consider arrangements of hard-core parallel squares in a fixed rectangular box, where each square has to rest on either two squares below it or on the box's floor, and we put a uniform probability distribution on the set of all such arrangements. Then we run Markov chain Monte Carlo simulations and measure the packing fraction of the Markov chain configurations.

We begin more ambitiously by discussing a more realistic model. As is standard in Edwards theory we take as a starting point a variant of the hard sphere model of equilibrium statistical mechanics. Consider a model consisting of large collections of impenetrable, unit mass, unit diameter spheres in a large container, acted on by gravity and with infinite coefficient of friction between themselves and with the container. Put a probability density on the set of all mechanically stable packings of the spheres in their container, with the probability density of a packing c proportional to $\exp[-E(c)]$, where $E(c)$ is the sum of the heights, from the floor of the container, of the centers of the spheres in the packing c. We expect, but cannot show, that such an ensemble will exhibit a gradient in the volume fraction (with volume fraction decreasing with height) and that there is a well-defined random loose packing density as one approaches the top of the packing (where the analogue of hydrostatic pressure goes to zero). By a "well-defined random loose packing density" we mean that as one takes an infinite volume limit, the probability distribution for the volume fraction of the top layer of the packing becomes concentrated at a single nonzero value. We emphasize that we are focusing on a bulk property near the top of the configuration, not a surface phenomenon.

The above determines a well defined zero pressure probability distribution for packings c. One could imagine simulating the distribution with Monte Carlo or molecular dynamics, but this is not practical at the high densities which are necessary in a granular model. (We emphasize that any such simulation should reproduce the above probability distribution; in this Edwards-style model the equilibrium probability distribution is completely determined, so there is no freedom available in deciding how packings are simulated.)

To make Monte Carlo simulations feasible, we make several simplifications in the way gravity and friction are incorporated in the above model. First we switch to an ensemble consisting of packings which are limits, as the gravitational constant goes to zero, of mechanically stable packings; we effect this by setting $E(c) = 0$ in the relative density $\exp[-E(c)]$. With this simplification configurations are now, in their entirety, representative of the top layer in the original model. Next we consider the two dimensional version of the above: congruent frictional unit disks in mechanically stable configurations under vanishingly small gravity. Note that each such disk must be in contact with either a pair of supporting disks below it or part of the container. (Here and elsewhere in this paper we neglect events of probability zero, such as one sphere perfectly balanced on another.) We simplify the role of gravity and friction in the model one last time by replacing the disks by congruent squares, with edges aligned with the sides of the (rectangular) container, each square in contact with either a pair of supporting squares below it or the floor of the container. This is now a granular version of the old model of "(equilibrium) hard squares" [8], which is a simplification of "hard disks" and "hard spheres" (see [1] for a review), in which gravity and friction is neglected but kinetic energy plays a significant role. We emphasize that in our granular model there is no longer any need to concentrate on the "top layer"; in fact we will eventually be concerned with an infinite volume limit which, as usual, focuses on the middle of the collection of squares and lets the boundaries grow to infinity. (We note that the model is capable of handling higher densities by constraining the squares to lie in a tightly containing box. We also note recent work by Song et al. [20, 22] which takes a different path, employing a mean field approximation instead of a simplified short range model which can be fully simulated, as we have done.)

We have run Markov chain Monte Carlo simulations on this model with the following results. We initialize the squares in an allowed configuration of some well-defined volume fraction anywhere between 0.5 and 1. If the initial volume fraction ϕ is not approximately 0.76, the simulation gradually expands or contracts the packings until the packing fraction reaches the range 0.76 ± 0.01; see Figs. 1 and 2. Furthermore, as the size of the packings increases, the standard deviation of the volume fraction tends towards zero.

The process is insensitive to the dimensions of the containing box except for extremes. We choose the height of the box to be large enough so that the configurations of squares cannot reach the ceiling (so the box height becomes irrelevant). We must choose the box width more carefully, since if the side walls of the containing box abut a closely-packed initial configuration, the simulation cannot significantly change the volume fraction; alternatively, if the width of the box is much larger than that of the initial configuration, the simulation will produce a monolayer on the floor. We ignore both extremes, however, and find that the equilibrium volume fraction is otherwise insensitive to the width of the box. More precisely, we found that the equilibrium volume fraction should be accurate if the box width is between $2\sqrt{N}$ and $8\sqrt{N}$, where $N \geq 100$ is the number of squares. To understand these limits, first note that since we will be conjecturing the behavior of the model in the infinite volume limit, the equilibrium configuration should be a single bulk pile, so the box width should be on the order of $\sqrt{N}$. Regarding the lower bound, note that at any volume fraction a configuration occupies the least amount of floor space when the squares are arranged in a

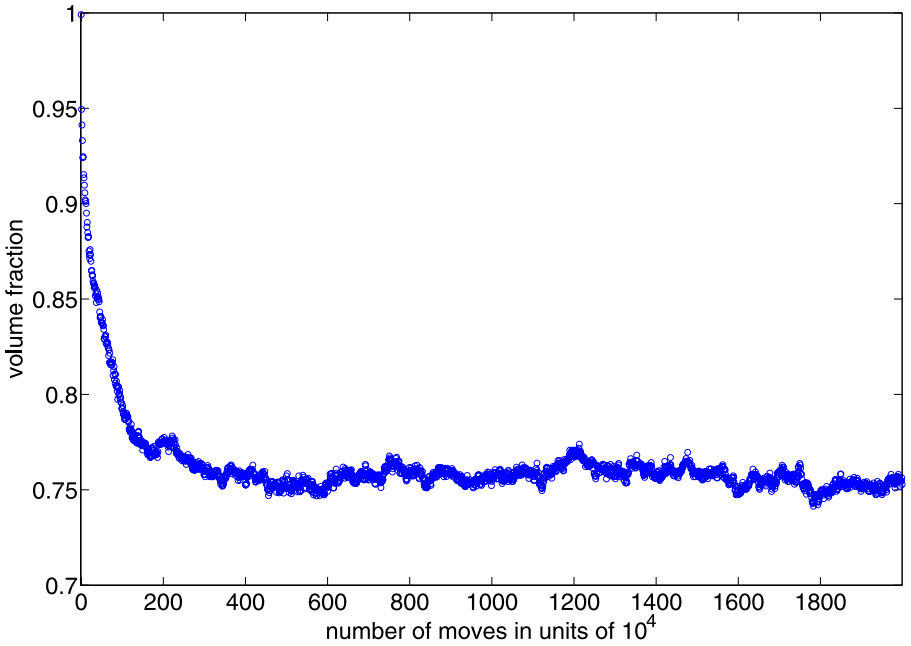


Fig. 1 Plot of volume fraction versus number of moves, from an initial volume fraction of 0.9991, for 970 squares

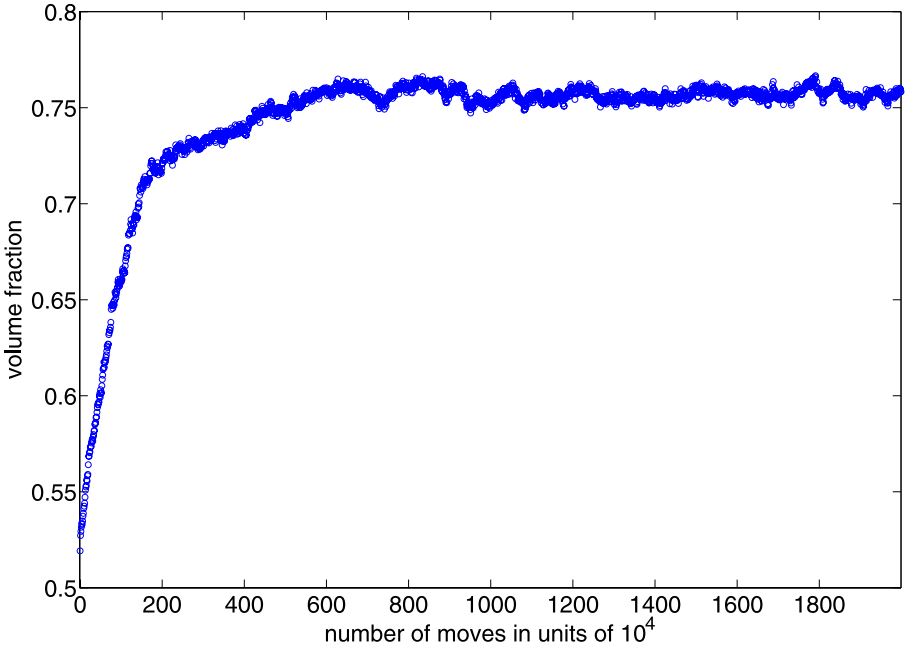


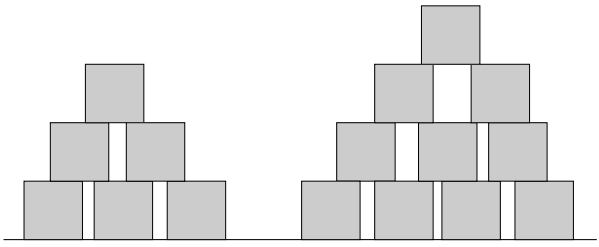
Fig. 2 Plot of volume fraction versus number of moves, from an initial volume fraction of 0.5192, for 994 squares

single full triangle. The bottom level of such a triangle has just under $\sqrt{2N}$ squares. Assume the containing box fits tightly around the triangle; if the triangle has volume fraction greater than 0.754 then the configuration will not be able to decrease to this equilibrium volume fraction. We avoid this by ensuring that the box width is at least $2\sqrt{N} > (0.754)^{-1}\sqrt{2N}$. To arrive at the upper bound we performed simulations on fixed particle number and let the box width vary. We found that the equilibrium volume fraction was reliable so long as the box width was less than about $8\sqrt{N}$, at least for $N \geq 100$.

We conclude that, for box widths in the aforementioned acceptable range, the equilibrium volume fraction depends only on the number of squares in the system. The main goal of our work is an analysis of the distribution of volume fraction—both the mean and standard deviation—as the number of particles increases. We conclude that the limiting standard deviation as particle number goes to infinity is zero, so the model exhibits a sharp value for the random loose packing density, which we estimate to be approximately 0.754.

The heart of our argument is the degree to which we can demonstrate that in this model there is a sharp value, approximately 0.754, for the equilibrium volume fraction of large systems, and we postpone analysis of error bars to later sections. But to understand the value 0.754, consider the following crude estimate of the volume in phase space of all allowable packings at fixed volume fraction ϕ. First notice that the conditions defining the model prevent the possibility of any "holes" in a configuration. Furthermore, if we consider any rectangle in the interior of a configuration, each horizontal row in the rectangle contains the same number of squares. (One consequence is that in the infinite volume limit each individual configuration must have a sharply defined volume fraction; of course this says nothing about the width of the distribution of volume fraction over all configurations.) Now consider a very symmetrical configuration of squares at any desired volume fraction ϕ, with the squares in each horizontal row equally spaced, and gaps between squares each of size $(1-\phi)/\phi$ centered over squares in the next lower horizontal row; see Fig. 3. Consider these squares to represent average positions, fix all but one square in such a position, and consider the (horizontal) degree of motion allowed to the remaining square. There are two constraints on its movement: the gap size separating it from its two neighbors in its horizontal row, and the length to which its top edge and bottom edge intersects the squares in the horizontal rows above and below it. These two constraints are to opposite effect: increasing the gap size decreases the necessary support in the rows above and below. A simple calculation shows that the square has optimum allowed motion when the gap size is $1/3$, corresponding to a volume fraction of 0.75, roughly as found in the simulations. In other words, this argument suggests that the volume in phase space (which for N squares we estimate to be L^N, where L is the allowed degree of motion of one square considered above) is maximized among allowed packings of fixed volume fraction by the packings of volume fraction about 0.75. Note that this is only a free volume-type estimate, so it is by no means a proof that a sharp entropy-maximizing volume fraction exists or is equal to or near 0.75.

Fig. 3 An allowed configuration



To obtain accurate physical measurements a fluidization/sedimentation method has been developed to prepare samples of millions of grains in a controlled manner; see [9, 13] and references therein for the current state of the experimental data. In these experiments a fluidized bed of monodisperse grains sediment in a fluid. The sediment is of uniform volume fraction, at or above 0.55 depending on various experimental parameters. Recall that the old experiments of Scott et al. [17, 18] reported a value of 0.608 for ball bearings in air; to achieve the low value (0.55 ± 0.001 [9]) the grains need to have a high friction coefficient and the fluid needs to have mass density only slightly lower than the grains to minimize the destabilizing effect of gravity. (In the absence of gravity one could still produce a granular bed by pressure; we do not know of experiments reporting a random loose packing value for such an environment.)

Given the dependence of the lowest achievable density on the characteristics of the experiment, we need to clarify the goal of this paper. From the physical perspective it is interesting that, for any fixed coefficient of friction and fixed relative density between the grains and background fluid, there seems to be a sharply defined lowest volume fraction achievable by bulk manipulation. It is possible furthermore that by suitably varying the coefficient of friction and relative density there is a single lowest possible volume fraction (currently believed to be about 0.55 [9]); we expect that this is the case, and that this has a simple geometrical interpretation in terms of ensembles of frictional hard spheres under gravity, as discussed above. This was the motivation of this work, and it is supported by simulations of our model. Our results suggest that whatever the initial local volume fraction of the fluidized granular bed, on sedimentation (in low effective gravity) most samples would have a well-defined volume fraction, the random loose packing density, with *no intrinsic lower bound* on the standard deviation of the distribution of volume fraction.

There have been previous probabilistic interpretations of the random loose packing density, for instance [11, 15], as well as the recent mean field model of Song et al. [20, 22]. A distinguishing feature of our results is our analysis of the degree of sharpness of the basic notion, which, as we shall see below, requires unusual care in the treatment of error analysis.

In summary, we have performed Markov chain Monte Carlo simulations on a two dimensional model of low pressure granular matter of the general Edwards probabilistic type [4]. Our main result, superficially summarized in Fig. 8, is that in this model the standard deviation of the volume fraction decays to zero as the particle number increases, which indicates a well-defined random loose packing density for the model. This suggests that real granular matter exhibits sharply defined random loose packing; this could be verified by repeating sedimentation experiments [9] at a range of physical dimensions. Our argument is only convincing to the extent that the confidence intervals in Fig. 8 are small and justified, which required a statistical treatment of the data unusual in the physics literature. We hope that our detailed error analysis may be useful in other contexts.

2 Analysis of Simulations

We performed Markov chain Monte Carlo simulations on our granular model, which we now describe more precisely. We begin with a fixed number of unit edge squares contained in a large rectangular box B. A collection of squares is "allowed" if they do not overlap with positive area, their edges are parallel to those of the box B, and the lower edge of each square intersects either the floor of the box B or the upper edge of each of two other squares; see Fig. 3. Note that although the squares have continuous translational degrees of freedom in the horizontal direction, this is not in evidence in the vertical direction because of the stability condition: the squares inevitably appear at discrete horizontal "levels".

Markov chain simulations were performed as follows. In the rectangular container B a fixed number of squares are introduced in a simple "crystalline" configuration: squares are arranged equally spaced in horizontal rows, the spacing determined by a preassigned volume fraction ϕ, and with squares centered above the centers of the gaps in the row below it; see Fig. 4. The basic step in the simulation is the following. A square is chosen at random from the current configuration and all possible positions are determined to which it may be relocated and produce an allowed configuration. Note that if the chosen square supports a square above it then it can only be allowed a relatively small horizontal motion; otherwise it may be placed atop some pair of squares, or the floor. So the boundary of the configuration plays a crucial role in the ability of the chain to change the volume fraction. In any case the positions to which the chosen square may be moved constitute a union of intervals. A random point is selected from this union of intervals and the square is moved. The random movement of a random square is the basic element of the Markov chain. It is easy to see that this protocol is transitive and satisfies detailed balance, so the chain has the desired uniform probability distribution as its asymptotic state [12]. See Fig. 5 for a configuration of 399 squares after 10^6 moves. Our interest is in random loose packing, which occurs in the top (bulk) layer of a granular pile, and we assume that the entirety of each of our configurations represents this top layer. We emphasize that our protocol is not particularly appropriate for

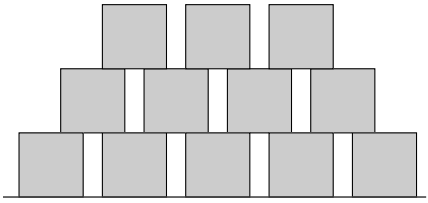


Fig. 4 A uniform configuration

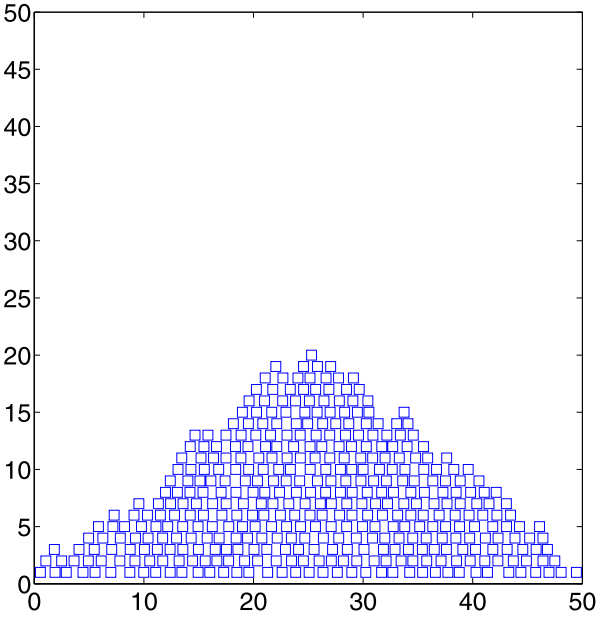


Fig. 5 399 squares after 10^6 moves

studying other questions such as the statistical shape of the boundary of a granular pile, or properties associated with high volume fraction, such as random close packing.

After a prescribed number of moves, a volume fraction is computed for the collection of squares as follows. Within horizontal level L_j, where $j = 0$ corresponds to the squares resting on the floor, the distances between the centers of neighboring squares is computed. (Such a distance is $1 + g$ where g is the gap between the squares.) Suppose that n_j of these neighboring distances are each less than 2, and that the sum of these distances in the level is s_j. At this point our procedure will be complicated by the desire to obtain information during the simulation about inhomogeneities in the collection, for later use in analyzing the approach to equilibrium. For this purpose we introduce a new parameter, p. For fixed $0 < p < 1$ we consider those levels, beginning from $j = 0$, for which n_j is at least $0.75p$ times the length of the box's floor. Suppose $L_{J(p)}$ is the highest level such that it, and all levels below it, satisfy the condition. We then assign the volume fraction

$$\phi(p) = \frac{\sum_{j=0}^{J(p)} n_j}{\sum_{j=0}^{J(p)} s_j} \tag{1}$$

to the assembly of squares. (The factor 0.75 represents the volume fraction we expect the box's floor to reach in equilibrium. Note that any two such calculations of volume fraction of the same configuration may only differ by a term proportional to the length of the boundary of the configuration, so any inhomogeneity is limited to this size.) Such a calculation of volume fraction was performed regularly, after approximately 10^6 moves, producing a time series of volume fractions ϕ_t for the given number of squares. (We suppress reference to the variable p for ease of reading. As will be seen later all our results correspond to the choice $p = 0.4$, so one can, without much loss, ignore other possible values.) Variables ϕ_t and ϕ_{t+1} are highly dependent, but we can be guaranteed that if the series is long enough then the sample mean:

$$\frac{1}{N} \sum_{t=1}^{N} \phi_t \tag{2}$$

will be a good approximation to the true mean of the target (uniform) probability distribution for the given number of squares [10].

We created such time series ϕ_t, each of about 10^4 terms (roughly 10^{10} elementary moves), using values $p = 0.2, 0.4, 0.6, 0.8$, on systems for the following numbers of squares: 100 m, and 1000 m, for $m = 1, 2, \ldots, 9$, with varying initial volume fractions. For each system we needed to determine the initialization period—the number of moves necessary to reach equilibrium—and then the total number of moves to be performed. Both of these determinations were made based on variants of the (sample) autocorrelation function $f(k)$ of the time series $\{\phi_t \mid 1 \le t \le T\}$ of volume fractions, defined for $1 \le k \le T$ by:

$$f(k) = \frac{1}{T - k + 1} \sum_{t=1}^{T-k+1} (\phi_t - \bar{\phi})(\phi_{t+k} - \bar{\phi}), \tag{3}$$

where $\bar{\phi}$ is the mean of the series. This function is easily seen to give less reliable results as k increases, because of limited data, so it is usual to work with functions made from it as follows. One way to avoid difficulties is to restrict the domain of f; we define the "unbiased" autocorrelation $f_1(k)$ by $f_1(k) = f(k)$ for $k \le T/10$. Another variant we consider is the

"biased" autocorrelation f_2, defined for all $1 \le k \le T$ by:

$$f_2(k) = \frac{1}{T} \sum_{t=1}^{T-k+1} (\phi_t - \bar{\phi})(\phi_{t+k} - \bar{\phi}), \tag{4}$$

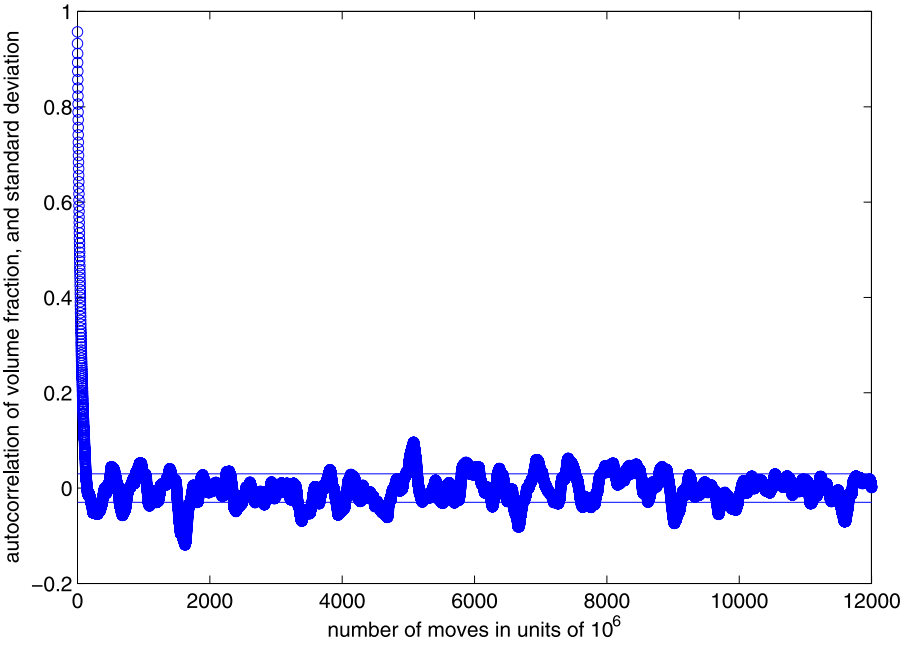
which reduces the value of $f(k)$ for large k. (See pages 321–324 of Priestley [14] for a discussion of this biased variant.) We consider both variants of autocorrelation; to refer to either we use the term f_j.

With these autocorrelations we determined the smallest $k = k_z$ such that $f_j(k) = 0$. We then computed the sample standard deviation σ_{f_j} away from zero of f_j restricted to $k \ge k_z$, and defined k_I to be the smallest k such that $|f_j(k)| \le \sigma_{f_j}$; see Fig. 6. (For ease of reading we sometimes do not add reference to j to quantities derived using f_j.) This defined the initialization period. Then starting from ϕ_{k_I} we recomputed the autocorrelation $\tilde{f}_j$ and $\sigma_{\tilde{f}_j}$ and determined the mixing time, the smallest $k = k_M$ such that $|\tilde{f}_j(k)| \le \sigma_{\tilde{f}_j}$. k_M was interpreted as the separation k needed such that the random variables ϕ_t and ϕ_{t+k} are roughly independent for all $t \ge k$. (We performed the above using the different definitions of volume fraction corresponding to different values of p, allowing us to analyze different geometrical regions of the samples. For each system of squares we selected, for initialization and mixing times, the largest obtained as above corresponding to the various values of p, which was always that for $p = 0.2$, corresponding to the lowest layers of the configuration.)

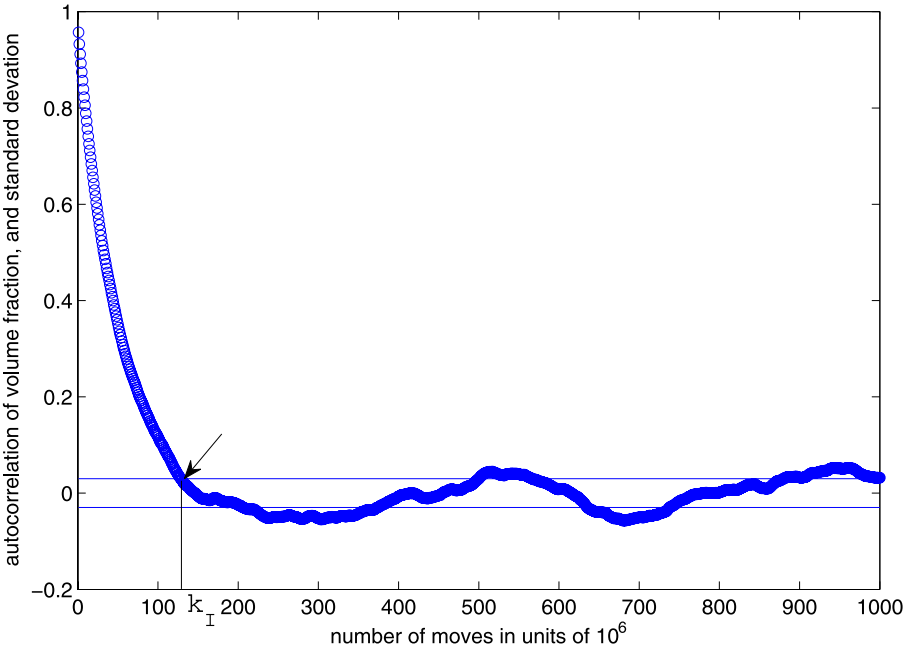
Once we determined k_I and k_M we ran the series to ϕ_F, where $F = k_I + Tk_M$ for some $T \ge 20$. The values of k_I and k_M are given in Tables 1 and 2; the empirical means and standard deviations of volume fraction are given in Tables 3 and 4.

In all our results we use $p = 0.4$ to minimize the boundary effects presumably associated with small or large p. (With large p the lowest level may have undue influence on the volume fraction; with small p, the surface levels could have undue influence. Note that the arrangements of squares on the lowest level and the surface levels are not restricted by the arrangements of squares below and above them, respectively, and so the corresponding volume fractions are not bound to the logic, discussed above, which suggested that each level should equilibrate at a volume fraction of about 0.75. In spite of this, we found that using any series corresponding to p in the range $0.2 \le p \le 0.8$ generated a similar result.) For all systems the volume fraction quickly settles to the range 0.76 ± 0.01 and we can easily see from Table 4 that the empirical standard deviations decrease with increase of particle number. In Fig. 7 we plot the empirical standard deviations against particle number, and in Fig. 8 the data is replotted using logarithmic scales. In Fig. 8 we include the best least-squares fit to a straight line $y = ax + b$, obtaining $a = -0.5004$ and $b = -0.8052$. The corresponding curve is included in Fig. 7. Also included in both graphs are 90% confidence intervals for the true standard deviations, obtained as described in the next section, using f_2. (There was not enough data to obtain a confidence interval by this method for the system with 8995 squares.) The same data is reanalyzed in Figs. 9 and 10 with confidence intervals derived using f_1.

We use the close fit to the line in Figs. 8 or 10, corresponding to 33 data points in a range of particle number varying from 100 to 9000, to extend the agreement to arbitrarily large particle number, and therefore to claim that the standard deviation is zero in the infinite volume limit, or that there is a sharp value for the random loose packing density. The argument is supported from the theoretical side by noting the closeness of the slopes in Figs. 8 and 10 to $-1/2$. A slope of $-1/2$ would be expected if it were true that an equilibrium configuration of N squares could be partitioned into similar subblocks which are roughly



(a)



(b)

Fig. 6 **a** Plot of biased autocorrelation (for 8000 particles) versus number of moves, with *horizontal lines* denoting one standard deviation away from zero. **b** Close-up of the above plot, with initialization time

Table 1 Basics (using unbiased autocorrelation f_1)

Number of squares in packing	Number of moves in units of 10^9	Step size in units of 10^6 moves	k_I (init. time) in units of step size using f_1	k_M (mixing time) in units of step size using f_1	Run length in units of mixing time using f_1
99	0.4	0.2	1	1	1998
100	0.4	0.2	1	1	1998
195	0.4	0.2	1	1	1998
205	0.4	0.2	1	1	1998
294	0.4	0.2	5	5	398
294	0.4	0.2	4	4	498
399	2	0.2	10	10	998
413	2	0.2	9	9	1110
497	2	0.2	17	17	587
504	2	0.2	17	17	587
600	2	0.2	12	12	832
603	2	0.2	23	23	433
690	2	0.2	14	14	713
690	2	0.2	20	20	498
790	2	0.2	43	43	231
803	2	0.2	14	14	713
913	2	0.2	34	34	293
913	2	0.2	75	73	135
996	12	1	13	13	953
1001	12	1	16	16	755
1955	12	1	38	38	318
2980	12	1	44	44	277
3003	12	1	51	53	234
3933	12	1	64	63	193
4008	12	1	99	75	158
4995	12	1	193	174	68
5908	12	1	163	96	125
6030	12	1	143	143	84
7037	12	1	223	261	46
7161	12	1	222	181	48
8015	12	1	132	120	100
8991	12	1	283	287	41
8995	12	1	632	631	18

independent—a proposition which would not be surprising given a phase interpretation of granular media [16]. Verifying such independence might be of some independent interest but would require much more data and much longer running times.

This is our main result, since it shows how to make sense of a perfectly well-defined random loose packing density within a granular model of the standard Edwards' form.

Table 2 Basics (using biased autocorrelation f_2)

Number of squares in packing	Number of moves in units of 10^9	Step size in units of 10^6 moves	k_I (init. time) in units of step size using f_2	k_M (mixing time) in units of step size using f_2	Run length in units of mixing time using f_2
99	0.4	0.2	1	1	1998
100	0.4	0.2	3	2	998
195	0.4	0.2	1	1	1998
205	0.4	0.2	1	1	1998
294	0.4	0.2	5	5	398
294	0.4	0.2	4	4	498
399	2	0.2	12	12	832
413	2	0.2	11	11	908
497	2	0.2	20	20	498
504	2	0.2	19	19	525
600	2	0.2	12	12	832
603	2	0.2	23	23	433
690	2	0.2	14	15	665
690	2	0.2	25	20	498
790	2	0.2	43	44	226
803	2	0.2	15	14	713
913	2	0.2	36	38	262
913	2	0.2	77	75	132
996	12	1	18	18	687
1001	12	1	16	16	755
1955	12	1	38	38	318
2980	12	1	56	56	218
3003	12	1	51	54	230
3933	12	1	76	78	156
4008	12	1	101	88	135
4995	12	1	191	177	67
5908	12	1	179	100	120
6030	12	1	139	151	80
7037	12	1	220	290	41
7161	12	1	199	186	47
8015	12	1	131	126	95
8991	12	1	282	355	33
8995	12	1	565	609	19

As to the actual asymptotic value of the volume fraction in the limit of large systems, we assume that our simulations suffer from a surface error proportional to $\sqrt{N}$ for a system of N squares. The least-squares fit of a function of form $A + B/\sqrt{N}$ to the data (see Figs. 11 and 12) yields $A = 0.7541$, and the good fit suggests an (asymptotic) random loose packing density in our granular model of about 0.754.

Table 3 Volume fraction

Number of squares in packing	Sample value	End points of 95% confidence interval for true value, using f_1	End points of 95% confidence interval for true value, using f_2
99	0.7637	0.7637 ± 0.0007	0.7637 ± 0.0007
100	0.7631	0.7631 ± 0.0007	0.7631 ± 0.0007
195	0.7617	0.7617 ± 0.0005	0.7617 ± 0.0005
205	0.7608	0.7608 ± 0.0005	0.7608 ± 0.0005
294	0.7605	0.7605 ± 0.0004	0.7605 ± 0.0004
294	0.7598	0.7598 ± 0.0004	0.7598 ± 0.0004
399	0.7596	0.7596 ± 0.0002	0.7596 ± 0.0002
413	0.7593	0.7593 ± 0.0002	0.7593 ± 0.0002
497	0.7590	0.7590 ± 0.0002	0.7590 ± 0.0002
504	0.7590	0.7590 ± 0.0003	0.7590 ± 0.0002
600	0.7583	0.7583 ± 0.0002	0.7583 ± 0.0002
603	0.7588	0.7588 ± 0.0002	0.7588 ± 0.0002
690	0.7581	0.7581 ± 0.0002	0.7581 ± 0.0002
690	0.7583	0.7583 ± 0.0003	0.7583 ± 0.0003
790	0.7578	0.7578 ± 0.0003	0.7578 ± 0.0003
803	0.7579	0.7579 ± 0.0002	0.7579 ± 0.0002
913	0.7575	0.7575 ± 0.0002	0.7575 ± 0.0003
913	0.7578	0.7578 ± 0.0003	0.7578 ± 0.0003
996	0.7575	0.7575 ± 0.0001	0.7575 ± 0.0001
1001	0.7574	0.7574 ± 0.0001	0.7574 ± 0.0001
1955	0.7565	0.7565 ± 0.0002	0.7565 ± 0.0002
2980	0.7558	0.7558 ± 0.0002	0.7558 ± 0.0001
3003	0.7559	0.7559 ± 0.0002	0.7559 ± 0.0003
3933	0.7554	0.7554 ± 0.0002	0.7554 ± 0.0002
4008	0.7558	0.7558 ± 0.0003	0.7558 ± 0.0003
4995	0.7555	0.7555 ± 0.0004	0.7555 ± 0.0005
5908	0.7549	0.7549 ± 0.0003	0.7549 ± 0.0003
6030	0.7551	0.7551 ± 0.0004	0.7551 ± 0.0003
7037	0.7545	0.7545 ± 0.0005	0.7545 ± 0.0004
7161	0.7550	0.7550 ± 0.0005	0.7550 ± 0.0007
8015	0.7551	0.7551 ± 0.0004	0.7551 ± 0.0004
8991	0.7551	0.7551 ± 0.0004	0.7551 ± 0.0012
8995	0.7550	none	none

Our argument concerning asymptotically large systems depends on the fit of our standard deviation data to a curve, and the degree to which this fit is convincing depends on the confidence intervals associated with our simulations. In the next section we explain how we arrived at our confidence intervals.

Table 4 Standard deviation of volume fraction

Number of squares in packing	Sample value	End points of 95% confidence interval for true value, using f_1	End points of 95% confidence interval for true value, using f_2
99	0.0160	0.0160 ± 0.0005	0.0160 ± 0.0005
100	0.0157	0.0157 ± 0.0006	0.0157 ± 0.0006
195	0.0110	0.0110 ± 0.0004	0.0110 ± 0.0004
205	0.0108	0.0108 ± 0.0004	0.0108 ± 0.0004
294	0.0090	0.0090 ± 0.0003	0.0090 ± 0.0003
294	0.0090	0.0090 ± 0.0003	0.0090 ± 0.0003
399	0.0078	0.0078 ± 0.0001	0.0078 ± 0.0002
413	0.0076	0.0077 ± 0.0001	0.0077 ± 0.0001
497	0.0070	0.0070 ± 0.0001	0.0070 ± 0.0001
504	0.0069	0.0069 ± 0.0001	0.0069 ± 0.0001
600	0.0064	0.0064 ± 0.0001	0.0064 ± 0.0001
603	0.0064	0.0064 ± 0.0001	0.0064 ± 0.0001
690	0.0060	0.0061 ± 0.0002	0.0060 ± 0.0001
690	0.0060	0.0060 ± 0.0002	0.0060 ± 0.0001
790	0.0055	0.0055 ± 0.0002	0.0055 ± 0.0002
803	0.0055	0.0055 ± 0.0002	0.0055 ± 0.0002
913	0.0052	0.0052 ± 0.0002	0.0052 ± 0.0002
913	0.0053	0.0053 ± 0.0001	0.0053 ± 0.0001
996	0.0050	0.0050 ± 0.0001	0.0050 ± 0.0001
1001	0.0049	0.0049 ± 0.0001	0.0049 ± 0.0001
1955	0.0036	0.0036 ± 0.0001	0.0036 ± 0.0001
2980	0.0028	0.0028 ± 0.0001	0.0028 ± 0.0001
3003	0.0029	0.0029 ± 0.0001	0.0029 ± 0.0001
3933	0.0024	0.0025 ± 0.0001	0.0025 ± 0.0001
4008	0.0025	0.0025 ± 0.0001	0.0025 ± 0.0002
4995	0.0022	0.0022 ± 0.0002	0.0022 ± 0.0002
5908	0.0021	0.0021 ± 0.0002	0.0021 ± 0.0002
6030	0.0020	0.0020 ± 0.0002	0.0020 ± 0.0002
7037	0.0018	0.0019 ± 0.0002	0.0019 ± 0.0002
7161	0.0019	0.0020 ± 0.0003	0.0021 ± 0.0004
8015	0.0016	0.0017 ± 0.0002	0.0017 ± 0.0002
8991	0.0017	0.0017 ± 0.0003	0.0021 ± 0.0008
8995	0.0016	none	none

3 Data Analysis

A good source for common ways to analyze the data in Markov chain Monte Carlo simulation is Chap. 3 in Newman and Barkema [12]. We will give a more detailed analysis, following the paper by Geyer [6] in the series put together for this purpose by the statis-

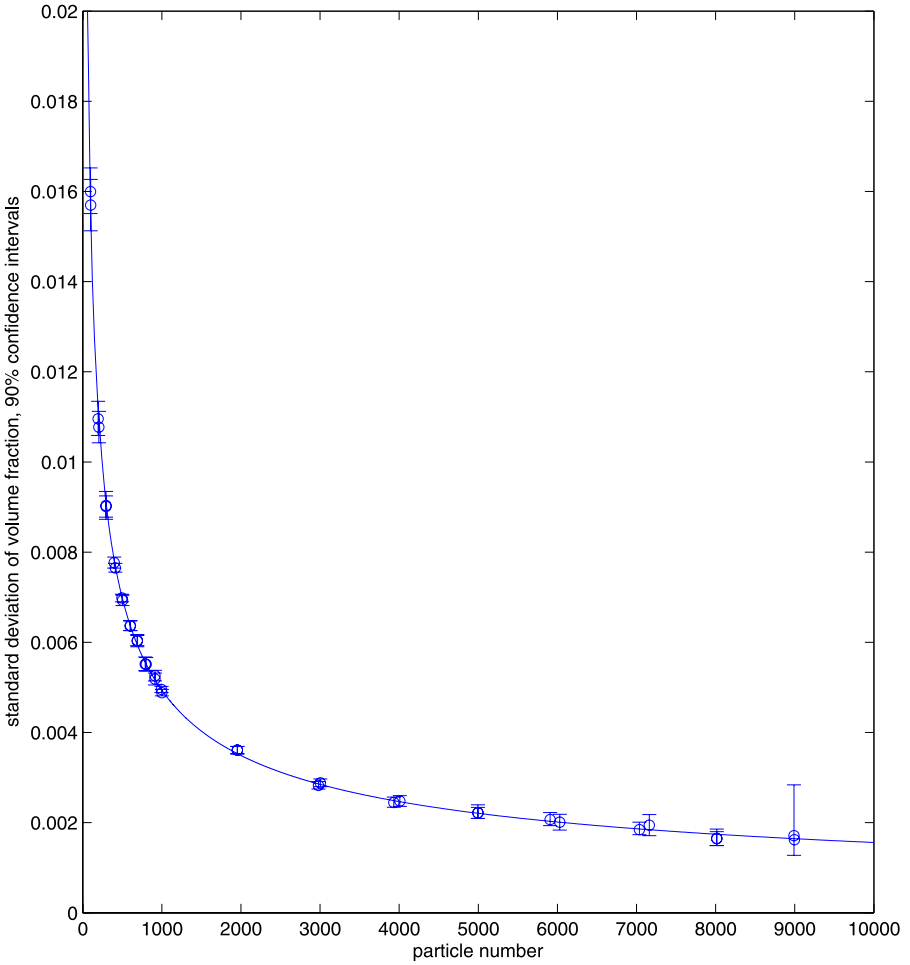


Fig. 7 Plot of the standard deviation of the volume fraction versus number of squares, using f_2 for confidence intervals

tics community [7]. As will be seen, our argument is based on the precision of estimates of various statistical quantities, and necessitates a delicate treatment.

Our simulations produce a time series c_j of (dependent) random configurations of squares. From this we produce other series $g(c_j)$ using functions g on the space of possible configurations c, in particular the volume fraction $g_1(c) = \phi$ and $g_2(c) = (\phi - K)^2$ for constant K.

We use the common method of batch means. As described in the previous section, we first determine an initialization time k_I and a mixing time k_M for our series c_j from autocorrelations. After removing the initialization portion of the series, we break up the remaining W terms of the series into $w \geq 2$ equal size consecutive batches (subintervals), each of the same length W/w, discarding the last few terms from the series if w does not divide W evenly.

It should be emphasized that rarely, if ever, are conclusions drawn from a finite number of Monte Carlo simulations a literal proof of anything interesting. We are going to obtain

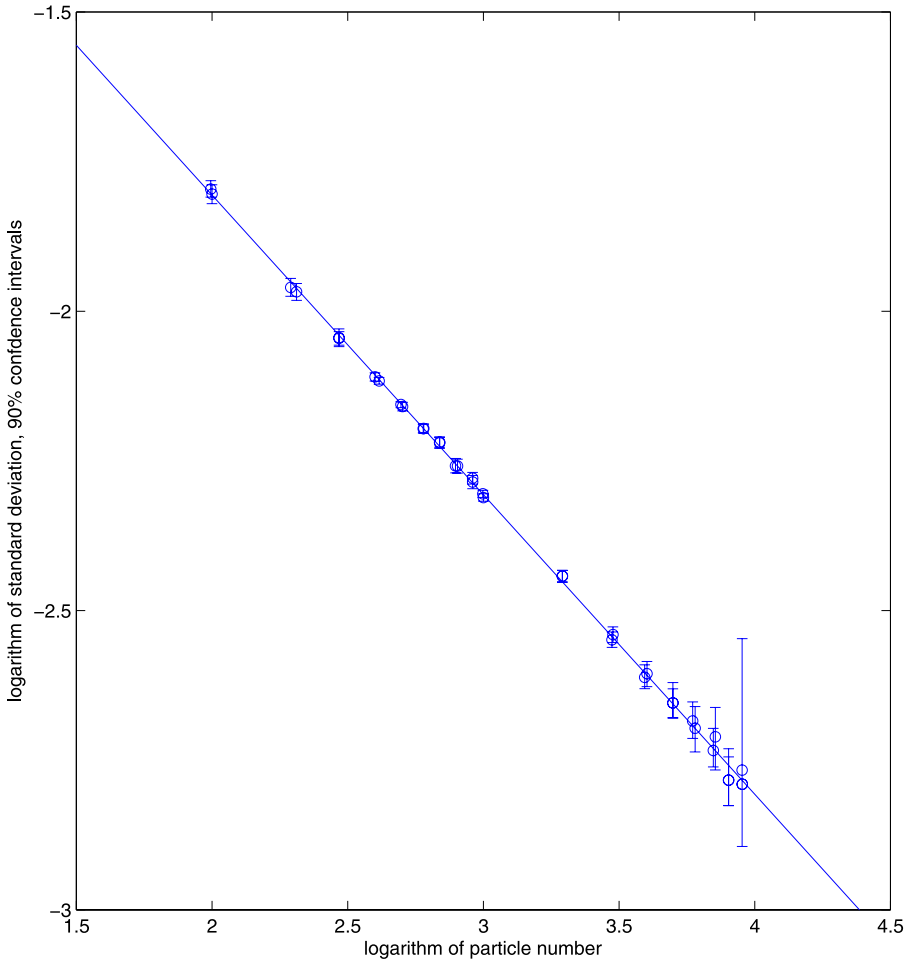


Fig. 8 Plot of the standard deviation of the volume fraction versus number of squares, using log scales and f_2 for confidence intervals. The line is $y = -0.5004x - 0.8052$

confidence intervals (using the Student's t-test) for the mean and standard deviation of the volume fraction of our systems of fixed particle number. The t-test's results would be mathematically rigorous if in our simulations we had performed infinitely many moves; of course this is impossible, so we will try to make a convincing case that we have enough data to give reliable results. Ultimately, this is the most sensitive point in our argument.

Assume fixed some function g, and denote the true mean of $g(c)$ by μ_g. Assume, temporarily, that enough moves have been taken for the t-test to be reliable. (We will come back to this assumption below.) With the notation $\overline{g(c)}$ for the empirical average $(1/w)\sum_k \langle g(c)\rangle_k$ of $g(c)$, where $\langle g(c)\rangle_k$ is the empirical average of the k^{th} batch, the random variable:

$$\frac{\overline{g(c)} - \mu_g}{\sqrt{\frac{1}{w(w-1)}\sum_k (\langle g(c)\rangle_k - \overline{g(c)})^2}} \tag{5}$$

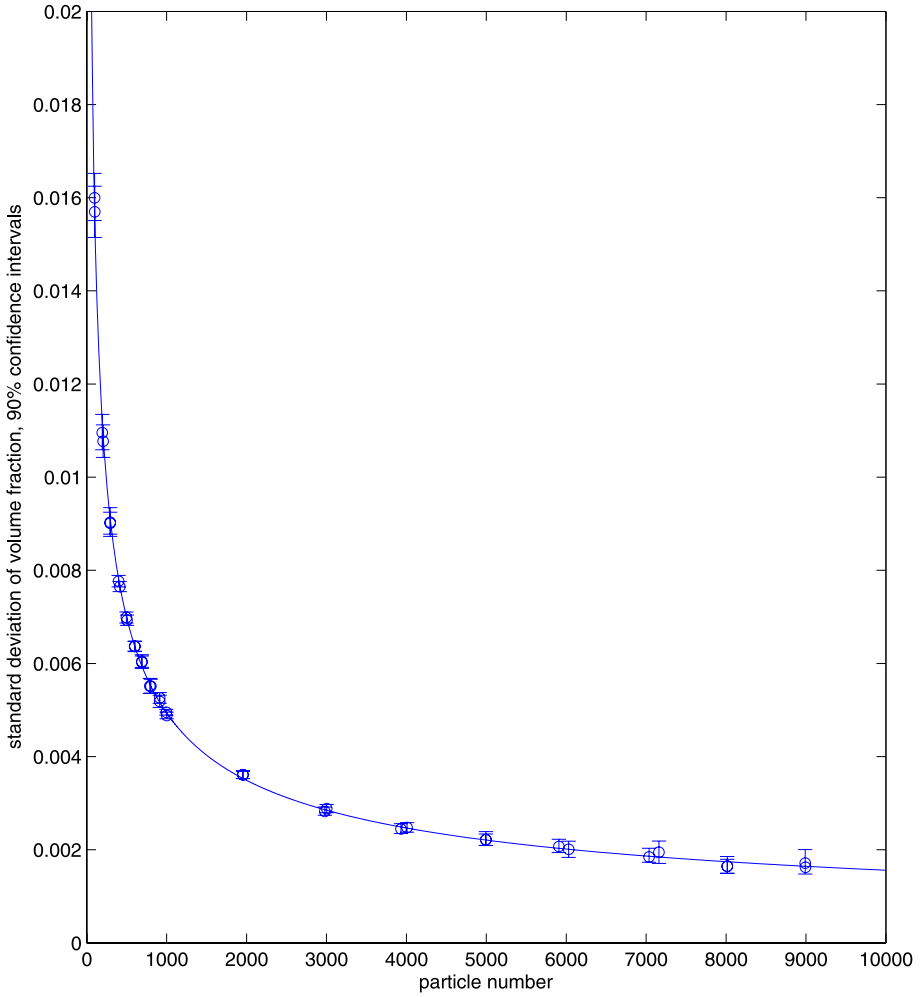


Fig. 9 Plot of the standard deviation of the volume fraction versus number of squares, using f_1 for confidence intervals

approximates a t-distribution, allowing one to compute confidence intervals for μ_g.

The above outline explains how (given the validity of the t-test) we could compute confidence intervals for the mean value of the volume fraction for the time series associated with our simulations for fixed numbers of squares. A small variation allows us to give confidence intervals for the standard deviations of these variables, as follows.

Denote the true standard deviation of $g(c)$ by σ_g. Using conditioning,

$$\begin{aligned}
&\text{Prob}(\mu_g \in I \text{ and } \sigma_g \in J) \\
&\quad = \text{Prob}(\mu_g \in I)\text{Prob}(\sigma_g \in J \mid \mu_g \in I) \\
&\quad = \text{Prob}(\mu_g \in I) \sum_i \text{Prob}(\sigma_g \in J \mid \mu_g \in I_i)\text{Prob}(\mu_g \in I_i \mid \mu_g \in I), \qquad (6)
\end{aligned}$$

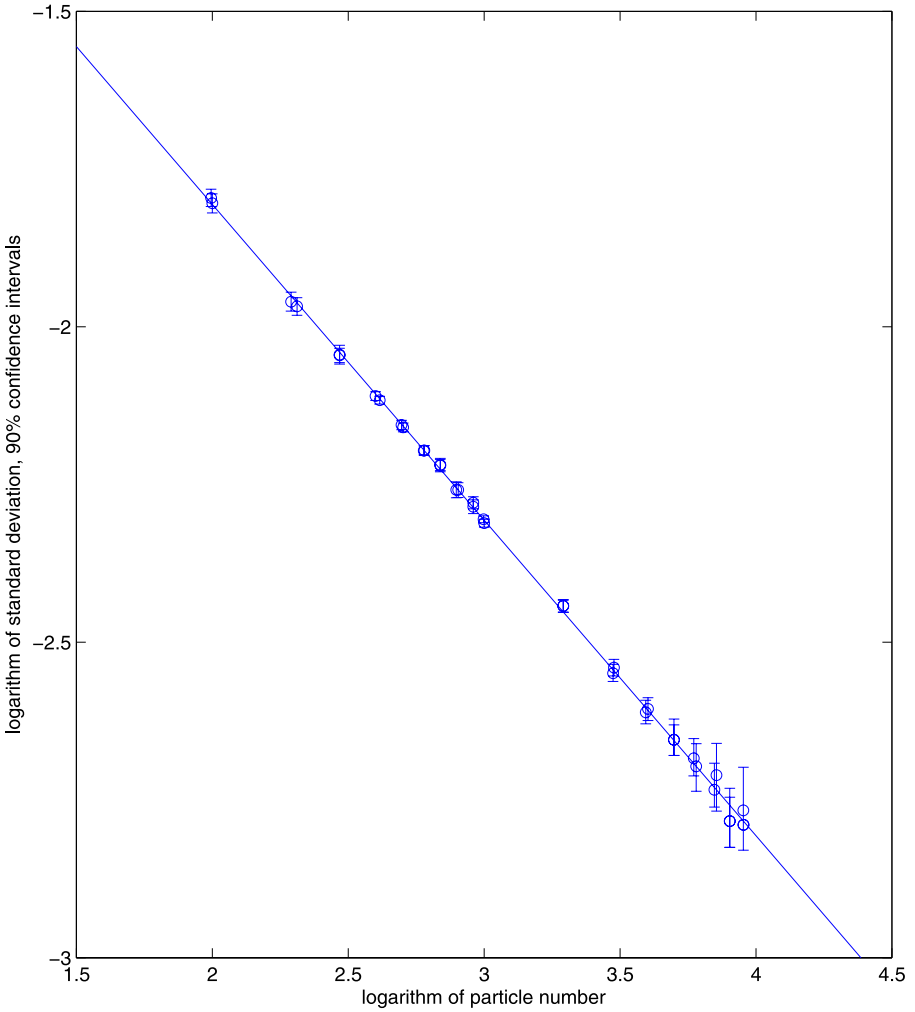


Fig. 10 Plot of the standard deviation of the volume fraction versus number of squares, using log scales and f_1 for confidence intervals. The line is $y = -0.5003x - 0.8055$

where $\{I_i\}$ is a partition of I. We have discussed how to obtain I so that the factor $\text{Prob}(\mu_g \in I)$ is at least 0.95. We now want to obtain J so that the factor $\text{Prob}(\sigma_g \in J \mid \mu_g \in I)$ is also at least 0.95, and therefore $\text{Prob}(\mu_g \in I \text{ and } \sigma_g \in J)$ is at least $(0.95)(0.95) > 0.90$.

Consider, for each constant K, the random variable

$$\Sigma_K = \sqrt{(1/w)\sum_j [\langle g(c)\rangle_j - K]^2}. \tag{7}$$

Using (5) with $(g(c) - K)^2$ playing the role of $g(c)$, we can obtain a 95% confidence interval for the mean of Σ_K^2, which we translate into a 95% confidence interval J_K for the mean of Σ_K. Assume the partition so fine that within the desired precision $J_K = J_i$ only depends

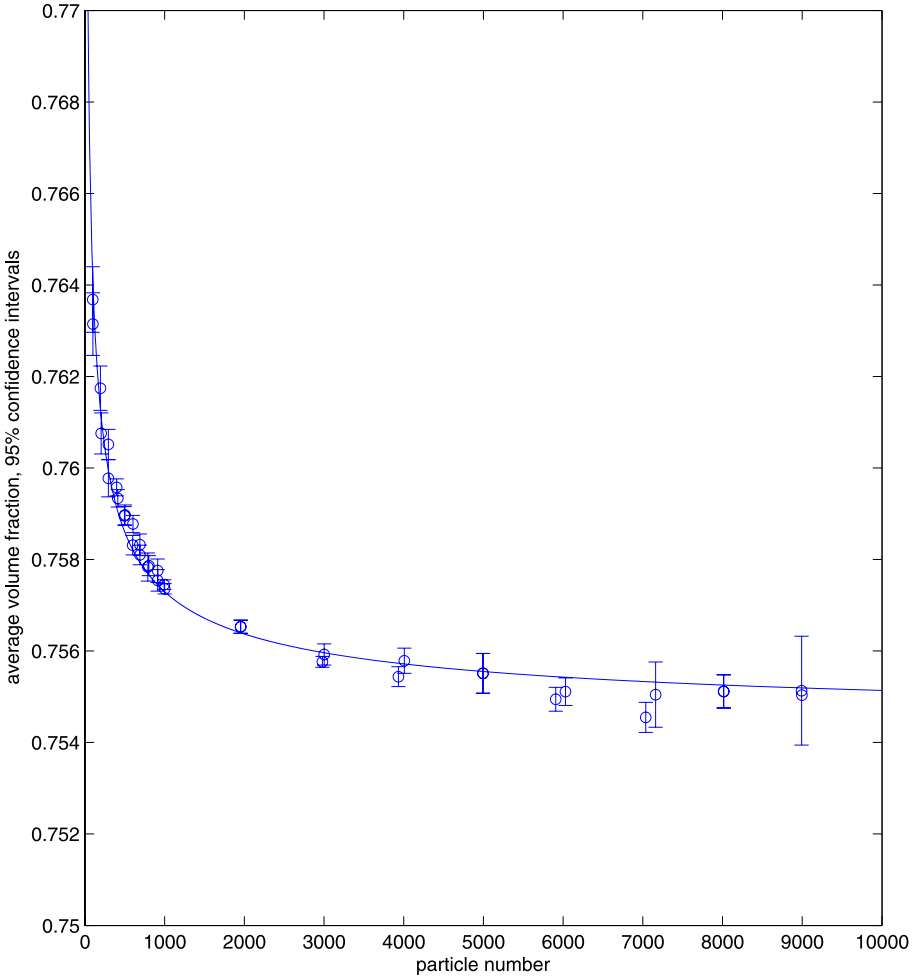


Fig. 11 Plot of the mean of the volume fraction versus number of squares, using f_2 for confidence intervals. The curve is $y = 0.7541 + 0.0998x^{-1/2}$

on i, where $K \in I_i$. Note that if $K = \mu_g$, then the random variable Σ_K has as its mean the standard deviation σ_g. So if we let $J = \bigcup_i J_i$, then $\text{Prob}(\sigma_g \in J \mid \mu_g \in I_i) > 0.95$ for all i, and therefore $\text{Prob}(\sigma_g \in J \mid \mu_g \in I) > 0.95$. In practice the union $J = \bigcup_i J_i$ is easy to compute.

In the above arguments we have assumed that enough moves have been taken to justify the t-test, which has independence and normality assumptions which are not strictly satisfied in our situation. We now consider how to deal with this situation. Some guidance concerning independence can be obtained from the following toy model.

Assume that for the time series of the simulation one can determine some number k_M, perhaps but not necessarily derived as above from the autocorrelation $f(k)$, such that variables ϕ_i and ϕ_{i+k} in the time series are roughly independent if $k \geq k_M$. We model this transition between independent random variables as follows.

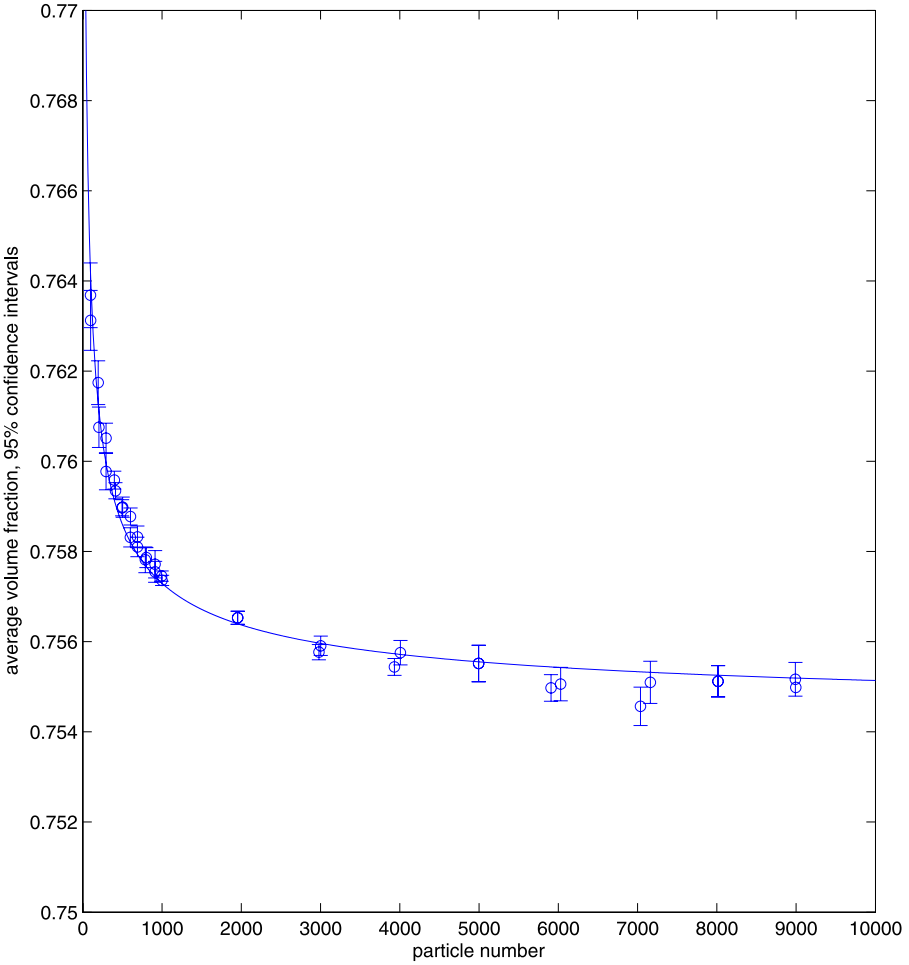


Fig. 12 Plot of the mean of the volume fraction versus number of squares, using f_1 for confidence intervals. The curve is $y = 0.7541 + 0.0998x^{-1/2}$

Let T and M be nonnegative (integer) constants. For $0 \le t \le T$ and $1 \le m \le M-1$ we first define independent, identically distributed random variables X_{tM} and from these define:

$$X_{tM+m} = \left(1 - \frac{m}{M}\right) X_{tM} + \frac{m}{M} X_{(t+1)M}, \tag{8}$$

together defining X_t for $0 \le t \le TM-1$. Note that variables X_t and X_{t+m} are independent for $m \ge 2M-1$.

A simple calculation shows that:

$$\sum_{m=0}^{M-1} X_{tM+m} = \left(\frac{M+1}{2}\right) X_{tM} + \left(\frac{M-1}{2}\right) X_{(t+1)M}. \tag{9}$$

Then another simple calculation shows that:

$$S_T \equiv \frac{1}{TM} \sum_{m=0}^{TM-1} X_m$$

$$= \frac{1}{T}\left[\sum_{t=1}^{T-1} X_{tM} + \frac{1}{2}(X_0 + X_{TM}) + \frac{1}{2M}(X_0 - X_{TM})\right]. \tag{10}$$

In other words S_T is the mean of roughly K *independent* variables.

Returning to the question of the assumptions in the t-test, the toy model suggests that the independence assumption is easily satisfied. The normality assumption is usually taken as the more serious [6]. But we note from [2] that the t-test is quite robust with respect to the normality assumption. Although the robustness of the t-test is well known and is generally relied on, in practice one still has to pick specific batch partitions in a reliable way. This is not covered in [6]. We arrived at a standard for batches of length 10 times mixing time for our series as follows. In outline, we use mixing times as computed above to standardize comparison between our systems with different particle number. Those for which our runs constituted at least 800 mixing times are assumed to give accurate values for the mean volume fraction. Various initial segments of these runs are then used, with various choices of batch partitions, to see which choices (if any) give reliable results for confidence intervals. Batches of size 10 mixing times proved reliable even for initial segments in the range of 20–100 mixing times, so this choice was then used for all systems. We emphasize that we are using this method to determine a minimum reliable batch size on the sequence of configurations, and then we apply this to the time series ϕ_t as well the time series $[\phi_t - K]^2$. We now give more details.

For most of the systems of particle numbers 100–900 we have over 500 mixing times worth of data, yet for some of the systems of particle numbers 1000–9000 we have, for practical reasons, less than one tenth that depth of data. We want to choose a fixed multiple of mixing time as batch length for all of our batches. To decide what range of mixing times will be reliable we used various portions of the data from those of our longest runs, and then applied the conclusions we drew to the other 3/4 of the runs.

More specifically, we treat as "reliable" the empirical volume fraction of the longest runs, those of length at least 800 times mixing time. We then consider a range of batch partitions of these systems to see which ones give accurate t-test results. We are looking for 95% confidence intervals, so we expect such intervals to contain the true volume fraction 95% of the time; since the true volume fraction is unknown we instead check how frequently the intervals contain the empirical volume fraction, which for the longer runs we have assumed is reliable. We do this for each of the runs of length 800 or more times mixing time. The results on these systems are the following.

For each of our longer runs (of at least 800 mixing times), we considered various initial portions of the run in each of six ranges of mixing times: 20–100, 100–200, 300–400, 400–500, and 500–600. For each of these truncated runs we considered batch partitions of the data into equal size batches of a variety of multiples of mixing time: 1–5, 6–10, 11–15, 16–20, 21–30, 31–40 and 41–50. For each size run and for each batch size we computed a 95% confidence interval for the true mean of the volume fraction, and determined whether or not the confidence interval covers the sample mean for the full run (which we are assuming is interchangeable with the true mean). The fraction of the more than 200 cases in each category for which the sample mean lies within the confidence interval is recorded in Table 5. From this it appears that using batches of size 1–5 mixing times would be unreliable,

Table 5 Fraction of times the given batch size gives acceptable confidence interval for given segment of total data of long runs, using unbiased autocorrelation f_1

		20–100 mixing times of total data	100–200 mixing times of total data	200–300 mixing times of total data	300–400 mixing times of total data	400–500 mixing times of total data	500–600 mixing times of total data
Number of	1–5	0.0849	0.0867	0.1119	0.1191	0.1938	0.2082
mixing	6–10	0.9410	0.9394	0.9830	1.0000	1.0000	1.0000
times	11–15	0.9648	0.9231	0.9656	1.0000	1.0000	1.0000
per batch	16–20	0.9524	0.9095	0.9777	0.9879	1.0000	1.0000
	21–31	0.9650	0.9177	0.9673	1.0000	1.0000	1.0000
	31–40	0.9712	0.9042	0.9643	1.0000	1.0000	0.9957
	41–51	0.9375	0.8869	0.9402	0.9511	0.9783	0.9402

Table 6 Fraction of times the given batch size gives acceptable confidence interval for given segment of total data of long runs, using biased autocorrelation f_2

		20–100 mixing times of total data	100–200 mixing times of total data	200–300 mixing times of total data	300–400 mixing times of total data	400–500 mixing times of total data	500–600 mixing times of total data
Number of	1–5	0.0833	0.0924	0.1182	0.1259	0.2006	0.2326
mixing	6–10	0.9245	0.9784	0.9805	0.9552	0.9762	1.0000
times	11–15	0.9107	0.9451	0.9607	0.9524	0.9707	1.0000
per batch	16–20	0.9728	0.9212	0.9745	0.9493	0.9655	1.0000
	21-31	0.9486	0.9059	0.9592	0.9547	0.9744	1.0000
	31–40	0.9780	0.9190	0.9592	0.9704	0.9852	1.0000
	41–51	0.9000	0.8639	0.9441	0.9565	0.9814	1.0000

but that size 10 times mixing times would be reliable. (Table 5 is based on mixing times obtained using the autocorrelation f_2. Table 6 is similar, using the autocorrelation f_1, and again justifies the use of batches of size 10 times mixing time.)

We then used batches of size 10 times mixing times to obtain 95% confidence intervals for the true mean of all the systems, obtaining the results tabulated in Table 3 and included in Figs. 11 and 12.

Finally, we applied the above batch criterion to obtain 90% confidence intervals for the true standard deviation of all our systems, using the method described earlier in this section. The results are in Table 4 and in Figs. 7 to 10.

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