Reply to "Comment on 'Jamming at zero temperature and zero applied stress: The epitome of disorder' "

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We answer the questions raised by Donev, Torquato, Stillinger, and Connelly (DTSC) in their preceding Comment on "Jamming at zero temperature and zero applied stress: The epitome of disorder" [Phys. Rev. E **70**, 043301 (2004)] We emphasize that we follow a fundamentally different approach than they have done to reinterpret random close packing in terms of the "maximally random jammed" framework. We define the "maximally random jammed packing fraction" to be where the largest number of initial states, chosen completely randomly, have relaxed final states at the jamming threshold in the thermodynamic limit. Thus, we focus on an ensemble of states at the jamming threshold, while DTSC are interested in determining the amount of order and degree of jamming for a particular configuration. We also argue that soft-particle systems are as "clean" as those using hard spheres for studying jammed packings and point out the benefits of using soft potentials.

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I. OVERVIEW: WHAT IS THE DIFFERENCE BETWEEN THE TWO APPROACHES?

The meaning of random close packing is fraught with ambiguities. A given amorphous packing can be made slightly more dense by introducing small amounts of crystallinity; thus, the concepts of "randomness" and "close-packing" would appear to be at odds with one another.

Torquato and co-workers [1] have pioneered a reexamination of random close packing (RCP) in terms of the notion of a "maximally random jammed" (MRJ) state, with specific definitions of "maximally random" and "jammed." The point of view espoused by Torquato *et al.* [1] and by Donev, Torquato, Stillinger, and Connelly [2] (DTSC) is fundamentally different from the one we have adopted [3,4]. They seek to identify, for a specific, finite configuration of hard spheres, the degree to which that configuration can be considered maximally random and jammed. They introduce three different categories of jammed states and employ a series of order parameters to measure the magnitude of different possible forms of order. A given configuration is maximally random if all of these order parameters are minimized with respect to variations of the particle positions and lattice vectors of the periodic cell. Thus, their emphasis is on finding the amount of order and degree of jamming in any given configuration.

By contrast, our point of view does not seek to identify the degree of order of any specific configuration. We are exclusively interested in defining an ensemble of states that are at the threshold of jamming. The results we have quoted were obtained by extrapolation to the thermodynamic limit not for infinitely hard spheres, but for soft particles that can overlap. In our studies, we have considered a configuration to be jammed if both the bulk and shear moduli are nonzero. One remarkable finding was that both of these moduli have their thresholds at the same packing density for all configurations that we studied. In addition, we examined the spectrum of vibrational modes and found that above the jamming threshold, all modes have nonzero frequency [5]. In our case, the "maximally random" density is defined in terms of an ensemble of configurations constructed as follows. Because we use soft particles, we can initially place N particles of volume v at random within a box of size L^d, where L is the box length and d is the dimensionality of space. (This corresponds to infinite temperature and cannot be done with hard spheres, which are never allowed to overlap.) Using a conjugate gradient or steepest descent algorithm, we relax the initial configuration at fixed packing fraction $\phi \equiv Nv/L^d$ to its nearest energy minimum; this defines the final state. This relaxation depends on the interparticle potential and not on any particle dynamics. It is therefore a property of the potential energy landscape. For a given number of particles, N, we define the "maximally random jammed *packing fraction*" to be where the highest number of initial states have final states at the jamming threshold. As we take the thermodynamic limit $N \to \infty$, we find that the width of the distribution of jamming thresholds approaches zero; this indicates that vir-

tually all of the configurations, *which were sampled randomly*, jam at the same packing fraction. The value of this packing fraction corresponds to the number commonly associated with random close packing. We verified that this distribution of jamming thresholds does not depend on the potentials we chose. Thus, while any given configuration can be jammed or not jammed, the "maximally random" density can be defined only by considering an ensemble of configurations.

The two approaches, that of DTSC and our own, are similar in that they reinterpret "random close packing" in terms of the "maximally random jammed" terminology [1]. We will argue that the two approaches are equally valid. In Sec. II, we respond to specific comments of DTSC. However, as we discuss in Sec. III, the question is not which of these approaches is more valid, but which is more useful.

II. RESPONSE TO SPECIFIC COMMENTS

A. What is "jammed"?

DTSC argue that we do not distinguish between the three different levels of jamming defined in Ref. [6]—namely "local," "collective," and "strict" jamming. It is indeed true that we are not interested in "local" jamming, the least restrictive of their definitions, in which groups of particles are free to move. Instead, we are interested in systems where the bulk and shear moduli are nonzero. By also ensuring that all the vibrational modes have positive frequency (in other words, that the dynamical matrix is positive definite so that configurations that are only locally jammed are excluded), we not only guarantee that the moduli are nonzero but also that the system is isostatic at the jamming threshold. In an isostatic system, the elastic properties are independent of the interparticle potential and thus dependent only on the geometry of the configuration. Thus, the soft-particle system is as "clean" as hard-sphere systems for studying the purely geometrical properties of the physical point *J*.

DTSC demonstrate in their Comment that our definition of "jamming" is closely related to their definition of "collective jamming." As we have said in the Overview, we are interested in the thermodynamic limit, when the number of particles in our system approaches infinity. In such a limit, boundary conditions no longer affect whether or not a system is jammed. Thus, the distinction between their definitions of "collective" and "strict" jamming disappears.

B. What is "random"?

In our framework, we have concentrated on creating a completely random set of configurations for the initial state. In our view, this is where "randomness" enters the problem. We thus find the fraction of all phase space that is funneled down (upon relaxation) into jammed configurations (i.e., the fraction of phase space that has inherent structures that are jammed). This sampling can easily be done with soft particles, but is impossible with hard spheres. For our systems, this provides a consistent and well-defined ensemble with which to work.

We agree with DTSC that we would also like to know the distribution of all possible final states at zero energy that are at the jamming threshold. If that knowledge were available, then it would be possible to define "maximally random jammed" with reference only to the final states. Unfortunately, an algorithm to find such a distribution is unavailable. Such an approach would be complementary to ours but would not supplant it.

We note that all of our *final* states at zero energy (i.e., states at or below the jamming threshold) are allowed hard-sphere states. Presumably this is why DTSC are particularly interested in the randomness of final configurations as opposed to initial ones. We can take the limit of using harder and harder potentials to see if any of our initial-state distributions change on approaching the hard-sphere limit. We used $V(r)=\epsilon\alpha^{-1}(1-r/\sigma)^{\alpha}$ for $r<\sigma$, $V(r)=0$ for $r\geqslant\sigma$, where σ is the particle diameter [7]. We find that the distributions are indistinguishable for three different values of α—namely, $5/2$, 2, and $3/2$. It is for this reason that we believe that our results are relevant to studies of hard-sphere systems. Of course, since our data are numerical and because there are different ways of taking the hard-sphere limit, including qualitatively different kinds of potentials, one can always worry that the results might change as one approaches the hard-sphere limit more closely.

In order to define the "maximally random jammed" density, we focus only on the distributions of configurations. From our point of view, randomness does not describe a particular configuration, but rather the ensemble of initial states. Contrary to the assertion of DTSC, we are not proposing a unique definition of order for the ensemble. Following their example of a jammed but diluted fcc lattice packing, this would be an allowed but *highly* improbable state in our distribution.

The example of a two-dimensional monodisperse disk packing is more problematic [8]. It is unclear for such a situation whether the phrases "maximally random jammed" or "random close packed" are appropriate. Indeed if one *only* had such a system one would never have come up with the idea of random close packing. However, our definition in terms of the ensemble would still yield a well-defined MRJ density. The identification of various types of order in a given packing is a deep and interesting question. However, it is unclear whether one wants to conflate that issue with a definition of an RCP or MRJ density. It depends on what one wants to learn from the definition.

C. Universal algorithms

Contrary to the assertion of DTSC, we are not claiming to explore the space of all jammed configurations in an unbiased manner. Rather, we are exploring the space of all initial states (at infinite temperature, $T=\infty$) in an unbiased manner. DTSC see no difference between starting at $T=\infty$ or any other temperature; the advantage of starting at $T=\infty$ is that one can at least sample initial states completely randomly.

We are glad that DTSC have pointed out something that may have been confusing in our paper. Because we have used two different protocols to determine different types of

results in our paper, they believe that we have mixed them up in determining the distribution of jamming thresholds, $P_j(\phi)$. This is not so. In determining $P_j(\phi)$ (Fig. 6 of Ref. [3]) we used the first protocol where we never varied the volume of our system during the relaxation process. We have said this explicitly in Sec. II C of our paper. To find the *distribution* of jamming thresholds, it is not necessary to determine the value of ϕ_c for any given *configuration*. Instead we only need to know the fraction of states, $f_j(\phi)$, that are jammed at any value of ϕ. $P_j(\phi)$ is then the derivative of $f_j(\phi)$ with respect to ϕ. In order to find $f_j(\phi)$ we need only determine if a state, produced by relaxation at a *fixed* packing fraction, is jammed or unjammed. This is, as we said earlier, easy to do since it only involves calculating whether the final configuration has a positive-definite pressure, shear modulus, or dynamical matrix. This knowledge, in and of itself, is sufficient to determine whether the state is jammed. Whether a configuration is jammed or not does not require knowledge of the precise value of ϕ_c for that state. We should point out that we could have obtained our results for the coincidence of the pressure and the shear modulus approaching zero at the same value of ϕ without ever using a compression or decompression run (the second protocol) but simply by plotting parametrically the pressure versus shear modulus for all states obtained by relaxation at fixed packing fraction (the first protocol).

DTSC objected that obtaining the distribution of jamming thresholds by looking at the fraction of jammed states is unphysical for hard spheres. Note that states that are at or below the jamming threshold are allowed hard-sphere packings. The fraction of such unjammed configurations $f_u(\phi)$, relevant to hard-sphere packings, is simply $1-f_j(\phi)$. Thus, the same distribution of jamming thresholds could have been obtained just as easily from the fraction of unjammed, hard-sphere, states.

III. WHY OUR APPROACH IS USEFUL

The question of which approach is more useful depends on what one wants to investigate. Perhaps the most important advantage of studying soft particles is that we can study properties of packings both above and below RCP or MRJ density. This allows a more complete picture of the properties of point *J*. For example, the divergence of the pair correlation function $g(r)$ near $r=\sigma$, $g(r)\sim(r-\sigma)^{-1/2}$ [3,4,9] was completely missed by studies of hard spheres, but was uncovered by using softer potentials. We have also found that the properties of a jammed configuration depend solely on $\phi-\phi_c$—that is, the distance from the jamming threshold (these studies were the only ones in which we allowed the density to vary, using the second procedure described by DTSC). Because we have shown that, at threshold, our states are isostatic, the mechanical properties of our system at ϕ_c (which approaches the RCP density in the infinite-system size limit) do not depend on the potential chosen but depend only on the geometry of the configuration. Thus, the soft-particle system is as "clean" as hard-sphere systems for studying the purely geometrical properties of point *J*.

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