Comment on "Classical Hard-Sphere Fluid in Infinitely Many Dimensions"

In a recent Letter[1] Frisch, Rivier, and Wyler (FRW) have derived what they describe as the exact equation of state of the fluid phase of the classical system of impenetrable D-dimensional spheres of *diameter a* in the limit $D \rightarrow \infty$. Their result is

$$p/(\rho k_B T) = 1 + (x/2), \tag{1}$$

where p is the pressure, $x = \rho v$, ρ is the number density, and v is called "the natural scale of volume" chosen equal to $V_a(D) = \pi^{D/2} a^D / \Gamma((D+2)/2)$, the volume of a D-dimensional spherical region of space of *radius a*. For the moment we defer scrutinizing the technical procedures which led to (1). Regarding (1) as given, the central issue is that this equation can be meaningful only for finite x. It is then imperative to ask, does the regime of finite x incorporate all densities where a fluid phase can be defined as the stable equilibrium phase and, additionally, as a metastable phase if such occurs? If not, Eq. (1) is incomplete and it fails to provide the exact equation of state of the hard-sphere fluid for $D \rightarrow \infty$. I claim that an unambiguous answer cannot be given to this question without resolution of two open problems. These are (a) the limiting behavior of the packing fraction $P(D)$ for the closet-lattice packing of impenetrable spheres, and (b) the largest density for which a fluid phase is defined in the large-D limit. Both problems are, in fact, outside the scope of the FRW treatment.

In any discussion of the equation of state of the fluid phase our preference is to express the pressure as a function of a physically relevant dimensionless variable constructed from the number density. The largest relevant density for a system of impenetrable D spheres surely cannot exceed $\rho_c(D)$, the density of closest-lattice packing. One is thus readily guided to the choice $y = \rho/\rho_c(D)$. The pair of dimensionless variables x and y are related by $x = 2M(D)y$, where $M(D) \equiv 2^{D-1}P(D)$. The exact values of $P(D)$ are known only for $D \leqslant 8$, but the Rogers lower bound and Blichfeldt upper bound on $P(D)$ yield[2] $D/e < M(D) < 2^{D-4}D$ for very large D. Far less is known about $y_{\max}$, the largest value of y for the fluid phase of an assembly of impenetrable D spheres. For $D = 2, 3, 4, 5$ the results of Monte Carlo molecular-dynamics (MCMD) simulations[3] suggest that a freezing transition occurs for $y \approx \frac{3}{4}, \frac{2}{3}, \frac{1}{2}, 0.4$. Suppose for the sake of the argument that one eschews the notion of a metastable fluid phase for densities exceeding that of the freezing transition, and suppose further that the very limited MCMD data lead one to speculate that $y_{\max}(D) \approx 3/(D+2)$ for all D. The largest value of the FRW variable is then $x_{\max}(D) = 2M(D) y_{\max}(D) \sim 6M(D)/D$ in the large-D limit. One thus arrives at the conclusion that if $y_{\max}(D) = O(1/D)$ then $x_{\max}(D)$ diverges in the large-D limit, and consequently Eq. (1) fails to provide the exact equation of state, unless the packing fraction is given by the Rogers lower bound. Alternatively, $x_{\max}(D)$ necessarily diverges if $y_{\max}(D)$ does not decrease to zero as fast as $1/D$.

The FRW result, Eq. (1), is obtainable from the standard Mayer theory[4] of nonideal gases by our adopting the approximation of truncating the rigorous expression connecting the cluster integral b_n and the irreducible integrals β_k by setting $\beta_k = 0$ for $k \geqslant 2$. That is, b_n is approximated as $n^{n-2}(\beta_1)^{n-1}/n!$. FRW state that this procedure is strictly valid in the large-D limit. It would appear that they base their claim on the argument that the single term retained in the expression for b_n dominates any individual discarded term by a factor which diverges as $D \rightarrow \infty$. Such an argument, however, leaves open the very real possibility that the combined contribution of all discarded terms is not ignorable in this limit. (The number of terms discarded from the exact expression for b_n is a very rapidly growing function of n.) A careful study of this issue would, in all likelihood, entail coping with the profound open problems described above. In any event, the claim that Eq. (1) describes the exact equation of state of the hard-sphere fluid for $D \rightarrow \infty$ is unfounded.

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[1] H. L. Frisch, N. Rivier, and D. Wyler, Phys. Rev. Lett. **54**, 2061 (1985).

[2] C. A. Rogers, *Packing and Covering* (Cambridge Univ. Press, Cambridge, 1964), pp. 3–4.

[3] For MCMD estimates of the freezing density of four- and five-dimensional systems of hard hyperspheres, see J. P. J. Michels and N. J. Trappeniers, Phys. Lett. **104A**, 425 (1984).

[4] See, for example, M. Toda, R. Kubo, and N. Saito, *Statistical Physics I* (Springer, New York, 1983), Eqs. (28), 29), (33), and (38) in Sec. 3.3.3.

