

Comment on “Novel Monte Carlo Approach to the Dynamics of Fluids: Single-Particle Diffusion, Correlation Functions, and Phase Ordering of Binary Fluids”

In their Letter [1] Kumar and Rao introduce a new technique for studying the late-time dynamics of a two-dimensional fluid. The approach consists of modeling the fluid by a fluctuating tethered network. Having described their method, Kumar and Rao go on to test their algorithm by computing the velocity autocorrelation function (VACF) for tagged particle motion in a system of hard disks.

Momentum is conserved in a fluid so local momentum cannot just disappear; part of it diffuses slowly away. As a result the VACF exhibits a slow algebraic decay—the so called “long-time tail” [2]. Since the 1970 paper by Alder and Wainwright [2], the VACF in a two-dimensional fluid is known to decay preasymptotically with a *positive* $1/t$ tail (the true asymptotic decay is slightly faster due to a “renormalization” effect [3,4]). By integrating the VACF over all times one obtains the diffusion coefficient. The Alder and Wainwright result implies that, in a two-dimensional fluid, the diffusion coefficient diverges to *plus* infinity (i.e., the mean squared displacement increases faster than linearly with time). Encouraged by successfully reproducing a $1/t$ decay of the VACF, the authors conclude that their model correctly describes the hydrodynamic behavior of real fluids and move on to study the late stages of spinodal decomposition.

However, the initial success is less than total as the tail in their VACF is *negative*. This is clearly illustrated in both Figs. 1 and 2 of Ref. [1]. Figure 1 of Ref. [1] shows a plot of the mean squared displacement divided by the time, as a function of time. This function (effectively a time-dependent diffusion coefficient) is decreasing with time, indicating that the VACF is negative. More explicitly, Fig. 2 of Ref. [1] shows a plot of the logarithm of *minus* the VACF, again indicating that the VACF is negative. Unless the authors are suggesting that the asymp-

totic decay of the VACF in a two-dimensional fluid really is negative, then, rather than being encouraged by their success, the authors should be concerned that their model does not properly reproduce the transport of momentum in a fluid. However, if they *are* suggesting that there is indeed a negative $1/t$ tail, then this not only contradicts all relevant theories and simulations, but predicts the unphysical phenomenon of a diffusion coefficient diverging to *minus* infinity (i.e., the mean *squared* displacement being negative). Clearly, the decay Kumar and Rao are observing is a transient and not the asymptotic decay. As the model used in Ref. [1] has no local momentum conservation we believe that it cannot reproduce the VACF found in a real fluid. In particular, hydrodynamic tails should be strongest at low viscosity, yet it is in precisely this regime that Kumar and Rao claim to observe *exponential* decay.

To conclude, the model proposed in Ref. [1] fails to account for an essential feature of the hydrodynamic behavior of real fluids. It is unclear to what extent this problem will have affected the observed long-time dynamics of spinodal decomposition studied in Ref. [1].

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- [1] P. B. S. Kumar and M. Rao, Phys. Rev. Lett. **77**, 1067 (1996).
- [2] B. J. Alder and T. E. Wainwright, Phys. Rev. A **1**, 18 (1970).
- [3] M. A. van der Hoef and D. Frenkel, Phys. Rev. Lett. **66**, 1591 (1991).
- [4] C. P. Lowe and D. Frenkel, Physica (Amsterdam) **220A**, 251 (1995).