Atomistic simulation of heat and mass transfer near the convection threshold

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The detailed behavior at the onset of Rayleigh-Bénard convection is examined using molecular-dynamics simulation of a hard-disk fluid in which the vertical temperature difference is increased gradually. The dependence of thermal flux and mass transfer rate on Rayleigh number just above onset is shown to be consistent with the assumption that the convective bifurcation is the analog of a second-order phase transition.

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Rayleigh-Bénard convection has long provided a framework for theoretical and experimental study of the spontaneous emergence of order in open dynamical systems [1–3]. While the phenomenon itself embraces a range of very distinct classes of behavior, from stationary rolls through various forms of time-dependent roll motion and eventually temporal chaos, possibly the most fascinating aspect is the initial bifurcation at which the system “decides” that convection provides a more efficient heat-transfer mechanism than conduction. The consequence of this decision is the spontaneous appearance of cooperative motion; the familiar roll patterns are the most prominent indication of the presence of long-range coherence. This dynamical analog of a thermodynamic phase transition, where the order parameter corresponds to the convective flow speed, is one of the simplest and most easily reproduced examples of self-organization in nature.

This Rapid Communication describes the results of a molecular-dynamics (MD) simulation of a two-dimensional Rayleigh-Bénard system as it is gradually driven from the conducting to the convecting state. The appearance of convection is accompanied by abrupt changes in the collective dynamics of the system: the measured heat and mass flows near the onset can be interpreted in the light of existing continuum theory. This represents the first attempt to use the discrete-particle MD approach to explore the details of dynamical transitions of this type.

MD simulation is widely used to study atomistic many-body problems by direct numerical solution of the classical equations of motion [4–6]; were it not for the MD approach the detailed dynamical behavior of such systems would be inaccessible. It has proved highly effective in the study of static and dynamic properties of equilibrium systems, and in the last few years has also proved able to deal with nonequilibrium problems, in particular those of a hydrodynamic nature such as fluid flow past an obstacle [7] and, of course, thermal convection [8–13]. While it is obvious that hydrodynamic behavior must eventually emerge from MD simulation, prior to these results there was little a priori expectation that systems with a mere $10^4$–$10^5$ particles can reproduce (with a satisfying degree of accuracy) effects normally observed over macroscopic length and time scales.

When applied to the thermal convection problem, MD simulation has displayed the development of well-formed convective roll structures [8, 9], the existence of time-dependent (in certain cases periodic) roll patterns [12, 13], and in some instances an acute sensitivity of the flow patterns to the finest details of the initial state [13]. Where quantitative comparisons have been carried out they have shown that the behavior follows the expectations of continuum hydrodynamics reasonably well [11–13]: examples include vertical temperature and density profiles, as well as the roll oscillation frequency.

The existence of a bifurcation at a particular value of the Rayleigh number (Ra) in an infinite horizontal fluid layer subject to a vertical temperature gradient is predicted by linear stability analysis of the Boussinesq form of the convection equations [14]; the actual critical value Ra_c depends on the properties of the thermal walls. Nonlinear analysis [1, 15] treats the steady-state behavior immediately following the onset of convection and leads to the functional dependence of convective flow speed and heat flux on the parameter $\epsilon = Ra - Ra_c$: flow speed and heat flux are proportional to $\epsilon^{1/2}$ and $\epsilon$, respectively. Such behavior can also be expressed in the language of second-order phase transitions [2] and the same $\epsilon$ dependence follows (using a mean-field treatment); the symmetry of the fluid that is broken at the transition is the flow field—below Ra_c the local flow is zero everywhere and the bifurcation at Ra_c is marked by the appearance of bulk flow. Theory is in excellent agreement with experiment for fluids such as oil and helium [2, 3], although the range of $\epsilon$ over which agreement holds turns out to be larger than expected.

There are two distinct approaches to MD computation [5]. The more widespread method is based on integrating the equations of motion by finite-difference methods and presumes a differentiable interparticle potential. The alternative is to use a step potential, in which case interactions between particles are of the impulsive type and the system evolves by means of a series of binary collisions [16]. In studies that emphasize qualitative behavior—the present work is an example—where the details of the potential function are not of great importance, convenience and computational efficiency dictate which method is preferable. For convection studies the step potential ap-
approach requires substantially less computational effort [13] and so the hard-disk fluid was used here.

In order to model thermal convection the standard MD approach must be supplemented by a suitable realization of the container walls and the inclusion of a uniform gravitational field. The bottom and top walls are responsible, respectively, for the introduction and removal of heat; this is accomplished by adjusting the velocities of particles as they collide with the walls in order to maintain the desired fluid temperature in the boundary layers. The walls themselves can be specified as either allowing or preventing slip at the boundaries; this work follows earlier simulations [13] and employs nonslip walls for greater realism. The effect of the gravitational field is that particle trajectories between impulsive collisions are parabolic rather than linear.

The system consists of 57600 hard disks packed in a square box at a number density of 0.4. The gravitational field strength g is chosen to maintain a balance between thermal and potential energy variations as the disks traverse the system vertically. With the exception of temperature the details are identical to [13].

The simulation is started with a uniform vertical temperature gradient imposed on the system; the lower and upper wall temperatures are $T_{\text{hot}} = 1.5$ and $T_{\text{cold}} = 1$ (the disks have unit mass and diameter, and the Boltzmann constant is also set to unity). Every 2000 time units $T_{\text{hot}}$ is increased by 0.5, and g is adjusted to maintain the energy balance. A single run of length $3.7 \times 10^9$ collisions was carried out (the number of collisions per temperature setting of course increases with $T_{\text{hot}}$); at the end of the run $T_{\text{hot}} = 9$, still below the constant value of 16 used in [13]. In terms of a characteristic time scale for the system, namely, the roll oscillation period (itself of the order of half the vertical thermal diffusion time), the time interval between temperature changes amounts to slightly less than half the oscillation period.

Heat transfer through the horizontal walls is computed directly at the instant of each particle-wall collision and the heat flux estimates are obtained by accumulating this data over measurement intervals of duration 100 time units. A total of 20 measurements are made at each $T_{\text{hot}}$ setting, but to allow the system to stabilize after each temperature change the first five are omitted from the analysis.

Coarse graining is used to smear out the discrete-particle nature of the fluid and permit the computation of space- and time-averaged quantities that can be analyzed as if they had emerged from a continuum fluid study. This approach permits flow streamlines as well as temperature and density profiles to be studied.

The most important raw data to emerge from the simulation are the time dependence of the heat and mass transfer rates. Figure 1 shows the heat flux (per unit wall length) divided by $\Delta T$ ($\Delta T = T_{\text{hot}} - T_{\text{cold}}$); if the transport coefficients are regarded as constant—an assumption that is not true in the present situation because of comparatively large temperature, density, and shear-rate variations across the system—then (heat flux)/$\Delta T$ is proportional to $Nu$, the Nusselt number. $Nu$ is defined as the ratio of total thermal flux to that due to

![FIG. 1. Time dependence of the heat flux over the entire run; the smoothed curve is intended to make the trends stand out from the fluctuations.](image1)

![FIG. 2. Time dependence of the maximum flow speed.](image2)

![FIG. 3. Flow streamlines on both sides of the convective bifurcation.](image3)
conduction alone. The solid curve shown on the graph is merely a running average of the scattered data points; the points themselves show the mean flux over intervals of 100 time units. Figure 2 shows the maximum coarse-grained cell flow speed; at the macroscopic level this is a measure of the convective mass transfer rate, but for microscopic systems there is a significant contribution from small-scale motion and thermal fluctuations.

The onset of convection near time 8000 is signaled by abrupt increases in both heat flux and flow speed. The streamlines in Fig. 3 display the change from disorganized localized flow patterns to a single container-filling roll; each such picture represents the mean flow over 100 time units (the average of 400 samples, each based on a 50 x 50 cell grid). The final single roll structure persists throughout the remainder of the computation, although near time 30000 a small roll appears in the upper left corner. (The time needed for this to develop should be contrasted with [13], where the typical requirement was 1000 time units.)

The only way to even approximately estimate Ra is based on Enskog theory for the transport coefficients [17] and an empirical equation of state [18]. While the computed value of Ra has absolutely no impact on the outcome of the simulation, it does provide some indication of the kind of behavior to be expected of the system if it is to resemble a macroscopic fluid. The result of the hard-disk analysis [11, 12] is \( \text{Ra}(\Theta) = b[(\Theta - 1)/(\Theta + 1)]^2 \), where \( \Theta = T_{\text{hot}}/T_{\text{cold}} \) and \( b \) is a constant (the limiting value of Ra as \( \Theta \to \infty \)). The difficulty in assessing the relevance of this result [12] is that the transport coefficients cannot be regarded as constant (neither spatially at any one temperature, nor as functions of time); such constancy is assumed in the Boussinesq analysis and is responsible for the central role of the Rayleigh number.

Figures 4 and 5 show the averaged data for (heat flux)/\( \Delta T \) and flow speed at each \( \Delta T \) value plotted against a reduced Ra based on the above form of Ra(\( \Theta \)) and assuming that \( b = 1 \). Error bars show the standard deviation of the grouped data points. Since the actual value of \( b \) obtained from the analysis is \( 7.8 \times 10^4 \), the onset of convection near Ra_c = 0.25 corresponds to an actual Rayleigh number of approximately \( 2 \times 10^4 \) (\( \Delta T = 2 \) at this stage of the simulation).

Superimposed on the two graphs are curves corresponding to the theoretically predicted functional forms that provide a visually reasonable fit to the simulation data near Ra_c. The linear \( \epsilon \) dependence of (heat flux)/\( \Delta T \) (or Nu) is shown in Fig. 4 (below Ra_c the value is assumed constant), and the \( \epsilon^{1/2} \) dependence of the convective flow in Fig. 5. No attempt has been made to refine these fits—it is not clear that either the quality of the data (contrast the experimental results [3]) or the degree of applicability of the theory warrant this.

The results overall are consistent with theory and experiment. It is hard to demand much better agreement when the coarse-grained cells typically contain as few as 20 particles, implying that the thermal velocity fluctuations are of comparable size to the convective flow speed. The deviations from Boussinesq behavior and the fact that the boundary layers extend a considerable distance into the fluid are also factors that can contribute to the deviations. Even without these complications the problem with small systems is a long-standing one: finite-size effects smear out the singularities in all numerical studies of critical phenomena.

While it still comes as a surprise that systems of the size considered here and in related work exhibit hydrodynamic behavior, there has been a gradual accumulation of evidence that such effects do appear in liquid simulations on length scales of only a hundred or so atomic diameters. Since systems of this size are now routinely studied by MD methods—at least in two dimensions—with computers becoming ever more powerful the opportunity for further exploration of the microscopic aspects of hydrodynamics is becoming increasingly feasible.