A recently introduced stochastic model for fluid flow can be made Galilean invariant by introducing a random shift of the computational grid before collisions. This grid shifting procedure accelerates momentum transfer between cells and leads to a collisional contribution to transport coefficients. By resumming the Green-Kubo relations derived in a previous paper, it is shown that this collisional contribution to the transport coefficients can be determined exactly. The resummed Green-Kubo relations also show that there are no mixed kinetic-collisional contributions to the transport coefficients. The leading correlation corrections to the transport coefficients are discussed, and explicit expressions for the transport coefficients are presented and compared with simulation data.

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Explicit GK relations for the transport coefficients of the SRD algorithm were derived in Ref. 8. In particular, it was shown that the shear viscosity, $\nu$, is given by

$$\nu = \frac{\tau}{Nk_BT} \sum_{n=0}^{\infty} \langle \sigma_{xy}(0)\sigma_{xy}(n\tau) \rangle,$$

(1)

where

$$\sigma_{xy}(n\tau) = -\frac{1}{\tau} \sum_j [v_{jx}(n\tau)\Delta \xi_{jy}(n\tau) + \Delta v_{jx}(n\tau)\Delta \xi^s_{jy}(n\tau)],$$

(2)

with $\Delta \xi_j(n\tau) = \xi_j([n+1]\tau) - \xi_j(n\tau)$, $\Delta \xi^s_j(n\tau) = \xi_j([n+1]\tau) - \xi^s_j([n+1]\tau)$, and $\Delta v_{jx}(n\tau) = v_{jx}([n+1]\tau) - v_{jx}(n\tau)$. $\xi_j$ is the cell coordinate of particle $j$ at time $n\tau$, while $\xi^s_j$ is its cell coordinate in the (stochastically) shifted frame. The prime on the sum indicates that the $t = 0$ term has the relative weight 1/2. The sum in Eq. (2) runs over all $N$ particles of the system. Here and in the following we have set the particle mass equal to one.

The straightforward evaluation of the GK relations presented in Ref. 8 leads to three contributions to the transport coefficients, which were called the kinetic, rotational, and mixed terms. For large mean free path, $\lambda = \tau \sqrt{k_BT} \gg a$, the assumption of molecular chaos is valid, and the kinetic contribution could be determined explicitly. For mean free paths smaller than the cell size, however, there are finite cell size corrections, and it was not possible to sum these contributions in a controlled fashion. The origin of the problem was the explicit appearance of the cell coordinate $\Delta \xi$ in the stress correlation functions.

In fact, the appearance of $\Delta \xi$ is troubling, since one would not expect this to be the case if the cell shifting procedure really does restore Galilean invariance. The key to resolving this dilemma is to realize that a proper resummation of the GK relations removes this dependence. In particular, by canceling $\xi$-dependent terms in successive contributions to the time series in Eq. (1) and using stationarity 12, it can be shown that transport coefficients are given by the same GK relations, but with the stress tensor $\sigma_{xy}(n\tau) \equiv \bar{\sigma}_{xy}^{kin}(n\tau) + \bar{\sigma}_{xy}^{rot}(n\tau)$, with

$$\bar{\sigma}_{xy}^{kin}(n\tau) = -\sum_j v_{jx}(n\tau)v_{jy}(n\tau),$$

(3)

and

$$\bar{\sigma}_{xy}^{rot}(n\tau) = -\frac{1}{\tau} \sum_j B_{jy}(n\tau)v_{jx}(n\tau),$$

(4)

where $B_{jy}(n\tau) = \xi^s_j([n+1]\tau) - \xi_j(n\tau) - \tau v_{jx}(n\tau)$. Note that the new stress tensor does not depend on $\xi$, the space-fixed cell coordinates of the particles. It can be shown 12, and has been verified numerically, that $\langle B_{\alpha} \rangle = 0$ and that all correlations of the $B$-fields with the particle velocities in the stress correlation functions factorize. Furthermore,

$$\langle B_{\alpha}(n\tau)B_{\beta}(m\tau) \rangle = \frac{a^2}{12} \delta_{\alpha\beta}(1 + \delta_{ij}) [2\delta_{n,m} - \delta_{n,m+1} - \delta_{n,m-1}],$$

(5)

so that the $B$'s are uncorrelated for time lags greater than one time step. These relations imply that there are only two—a pure kinetic and a pure rotational—contributions to the transport coefficients. Relation 5 is of central importance, because it contains all the geometrical features of the grid that contribute to the transport coefficients, and is independent of specific collision rules and particle properties.
the off-diagonal, and the triangles ($\boldsymbol{\Delta}$) the total contributions to a) the kinematic viscosity, $\nu_{\text{kin}}$, and b) the thermal diffusivity, $D_{T,\text{rot}}/a^2$, as a function of the collision angle $\alpha$. The bullets (●) are the diagonal, the squares (■) the off-diagonal, and the triangles (▲) the total contribution to the rotational viscosity and thermal diffusivity. The solid lines are the theoretical predictions. The data were obtained by time averaging over 360,000 iterations. Parameters: $L/a = 16$, $\lambda/a = 0.1$, $M = 3$ and $\tau = 1$.

Using these results in Fig. 1, the viscosity can be written as $\nu = \nu_{\text{kin}} + \nu_{\text{rot}}$, with

$$\nu_{\text{kin}} = \frac{\tau}{N k_B T} \sum_{n=0}^{\infty} \sum_{i,j=1}^{N} \langle v_{xi}(0)v_{yi}(0)v_{xj}(n\tau)v_{yj}(n\tau) \rangle \quad \text{(6)}$$

and

$$\nu_{\text{rot}} = \frac{\tau}{2N k_B T} \sum_{i,j=1}^{N} \{ \langle v_{xi}(0)v_{xj}(0) \rangle \langle B_{iy}(0)B_{yj}(0) \rangle + 2 \langle v_{xj}(0)v_{xj}(\tau) \rangle \langle B_{iy}(0)B_{yj}(\tau) \rangle \} \quad \text{(7)}$$

Assuming molecular chaos, it is straightforward to evaluate the kinetic contribution to the shear viscosity. If, in addition, it is assumed that the number of particles in any cell is Poisson distributed at each time step, with an average number $M$ of particles per cell, and average over the number of particles in a cell, one finds

$$\nu_{\text{kin}}^{2D} = k_B T \sum_{n=0}^{\infty} G_C(n\tau)$$

$$= \frac{k_B T}{2} \left[ \frac{M}{1 + e^{-M}} \sin^2(\alpha) - 1 \right], \quad \text{(8)}$$

in two dimensions, where

$$G_C(n\tau) \equiv \langle \delta_{xy}^{\text{kin}}(0)\delta_{xy}^{\text{kin}}(n\tau) \rangle_C/N(k_B T)^2$$

$$= \left[ 1 - 2 \sin^2(\alpha)(M - 1 + e^{-M})/M \right]^n. \quad \text{(9)}$$

The index $C$ indicates that molecular chaos was assumed when performing the averages. The corresponding result for Model A in three dimensions is

$$\nu_{\text{kin}}^{3D} = \frac{5M}{2} \left[ \frac{M}{1 + e^{-M}} \sin(\alpha) - \cos(\alpha) - \cos(2\alpha) \right] - 1$$

(see also Refs. [11, 12]).

Eqs. 5 and 10 are the same results one would obtain in the Chapman-Enskog approximation [1]. For small mean free path, however, there are significant contributions to $\nu_{\text{kin}}$ which are neglected in this approximation. They arise from correlations between particles which are in the same (shifted) cell at more than one time step. Fig. 1 contains a plot of the $G_C(n\tau)$ given in (9), and the open squares are simulation data for $\lambda/a = 1$. The agreement shows that for this value of the mean free path, Eq. 5 provides an excellent approximation for $\nu_{\text{kin}}$. On the other hand, data obtained for $\lambda/a = 0.01$ (●) exhibit much larger correlations for $n\tau \geq 2$. Fig. 2 contains a plot of the relative difference, $\delta G(2\tau) = G(2\tau) - G_C(2\tau)$ as a function of the mean free path. While it is rather difficult to evaluate these corrections analytically for general $\lambda$, we have calculated $\delta G(2\tau)$ in the $\lambda \to 0$ limit. The results of this calculation, which are shown in Fig. 2, are in excellent agreement with the numerical results.

There are corrections of this type at small $\lambda/a$ for all the transport coefficients, and it is important to note that they provide a particularly large contribution to the bare self-diffusion coefficient [4, 11, 12]. The effect of these corrections on the value of the viscosity is less significant and only visible at intermediate mean free path, since they vanish for large $\lambda$ and are small compared to the dominant collisional contribution for $\lambda \ll a$. For $\lambda/a = 0.4$, the correlations at $n = 2$ make an additional contribution of approximately 12% to the total viscosity.

The rotational contribution to the viscosity is easy to evaluate, since, as can be seen from Eq. 4, only stress correlation functions at equal time and for a time lag of one time step are required. Another simplifying feature is that because of momentum conservation, the diagonal (from $i = j$) and off-diagonal (from $i \neq j$) contributions to $\nu_{\text{rot}}$ in Fig. 1 obey the relation $\nu_{\text{rot}}^\text{diagonal} = 0$, since they vanish for large $\lambda$ and are small compared to the dominant collisional contribution for $\lambda \ll a$. For $\lambda/a = 0.4$, the correlations at $n = 2$ make an additional contribution of approximately 12% to the total viscosity.
Using this result and relation (3), and averaging over the number of particles in a cell, one obtains (12)

\[ \nu_{rot} = \frac{a^2}{6d \tau} \left( \frac{M - 1 + e^{-M}}{M} \right) [1 - \cos(\alpha)] , \]

for all the collision models we considered (the standard model in \( d = 2 \) and both models A and B in \( 3d \)) (11). Eq. (11) agrees with the result of Kikuchi et al (10) obtained using a different non-equilibrium approach in shear flow, but deviates slightly for small \( M \) from the result given in Refs. (9) and (11). Result (11) is compared with simulation results for the rotational contribution to the viscosity in Fig. 3.

The GK relation for the thermal diffusivity, \( D_T \), derived in Ref. (8) can be resumed in a similar fashion. In particular, it can then be shown that \( D_T = D_{T,kin} + D_{T,rot} \). \( D_{T,kin} \) was calculated in \( 2d \) in Ref. (9) and in \( 3d \) in (11) neglecting fluctuations in the number of particles in a cell. As for the viscosity, it is straightforward to include particle number fluctuations by averaging the contributions to the heat-flux correlation functions over the number of particles in a cell; the resulting expression will be given elsewhere (12). The relation \( D_{T,rot}^{\text{diagonal}} = -2D_{T,rot}^{\text{off-diagonal}} \), which follows from energy conservation, can be used to show that the rotational contribution to the thermal diffusivity is to leading order for large \( M \). Note that in contrast to the viscosity, the rotational contribution to the thermal diffusivity is \( O(1/M) \), so that the corrections to \( D_T \) at small \( \lambda/a \) arising from correlated collisions are more important than for the viscosity. Simulation results for \( D_{T,rot} \) are compared with Eq. (12) in Fig. 3.

It is now clear that the random shift procedure introduced in Refs. (7) and (8) not only restores Galilean invariance, but also enables an exact evaluation of the collisional contribution to the transport coefficients and clarifies several aspects of the underlying algorithm. In addition, the current approach justifies in detail several assumptions used in the non-equilibrium calculation of Kikuchi et al (11) which led them to the same, correct results for the shear viscosity derived here using GK relations. An advantage of the current approach is that it can be used to analyze the transport coefficients of the longitudinal modes, namely the bulk viscosity and thermal diffusivity, which are hard to calculate in a non-equilibrium approach (13). It can also be used to show that the bulk viscosity is equal to zero (8) (12).

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[1] A. Malevanets and R. Kapral, J. Chem. Phys. 110, 8605 (1999).
[2] A. Malevanets and R. Kapral, J. Chem. Phys. 112, 7260 (2000).
[3] Y. Inoue, Y. Chen, and H. Ohashi, J. Stat. Phys. 107, 85 (2002).
[4] A. Malevanets and J.M. Yeomans, Europhys. Lett. 52, 231 (2000).
[5] E. Falck, O. Punkkinen, I. Vattulainen, and T. Ala-Nissila, Phys. Rev. E 68, 050102(R) (2003).
[6] M. Ripoll, K. Mussawisade, R.G. Winkler, and G. Gompper, unpublished.
[7] T. Ihle and D.M. Kroll, Phys. Rev. E 63, 020201(R) (2001).
[8] T. Ihle and D.M. Kroll, Phys. Rev. E 67, 066705 (2003).
[9] T. Ihle and D.M. Kroll, Phys. Rev. E 67, 066706 (2003).
[10] N. Kikuchi, C.M. Pooley, J.F. Ryder, and J.M. Yeomans, J. Chem. Phys. 119, 6388 (2003).
[11] E. Tüzel, M. Strauss, T. Ihle and D.M. Kroll, unpublished.
[12] E. Tüzel, T. Ihle and D.M. Kroll, unpublished.
[13] Private communication, C.M. Pooley.