

Resummed Green-Kubo relations for a fluctuating fluid-particle model

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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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A recently introduced stochastic model for fluid flow [1,2] with efficient multiparticle interactions—which we will call Stochastic Rotation Dynamics (SRD)—is a promising tool for the coarse-grained modeling of a fluctuating solvent, particularly for colloidal [3] and polymer suspensions [4–6]. SRD can be thought of as a “hydrodynamic heat bath,” the details of which are not fully resolved, but which provides the correct hydrodynamic interactions between embedded particles. In addition to its numerical advantages, its simplicity makes it possible to obtain analytic expressions for the transport coefficients which are valid for both large and small mean free paths, something which is very difficult to do for other mesoscale particle-based methods.

In its original form [1,2], the SRD algorithm was not Galilean invariant at low temperatures, where the mean free path λ is smaller than the cell size a . However, as was shown in Refs. [7,8], Galilean invariance can be restored by introducing a random shift of the computational grid before every multiparticle interaction. A discrete-time projection operator technique was then used [8] to derive the Green-Kubo (GK) relations for the model’s transport coefficients. Using these results, explicit expressions for the transport coefficients were derived in an accompanying paper [9]. In particular, it was shown that the grid shifting procedure accelerates momentum transfer between cells and leads to a collisional contribution to the transport coefficients. However, the resulting expressions, while accurate, were only approximate, since it was not possible to sum up in any controlled fashion all the terms in the GK relations. Subsequently, Kikuchi *et al.* [10] used a nonequilibrium approach to derive expressions for the shear viscosity which differed slightly from those derived in [9]. Furthermore, while their approach yielded only two—pure kinetic and collision—contributions to the viscosity, the analysis of the GK formalism presented in Refs. [8,9] suggested that there are additional “mixed” contributions. These discrepancies led us to reexamine the GK approach.

In this paper we show that it is possible to resum the time series in the GK relation in such a way as to eliminate all dependence on the particles’ space-fixed cell coordinates. This leads to dramatic simplifications and allows the exact

evaluation of the collisional contribution to the transport coefficients. Furthermore, it is shown that there are only pure kinetic and collision contributions to the transport coefficients, with no cross terms.

In the SRD algorithm, the fluid is modeled by particles with continuous spatial coordinates $\mathbf{r}_i(t)$ and velocities $\mathbf{v}_i(t)$. The system is coarse grained into the cells of a regular lattice with no restriction on the number of particles in a cell. The evolution of the system consists of two steps: streaming and collision. In the streaming step, the coordinate of each particle is incremented by its displacement during the time step, τ . Collisions are modeled by a simultaneous stochastic rotation of the relative velocities (relative to the mean velocity of the particles in a cell) of *every* particle in each cell. As discussed in Refs. [7,8], a random shift of the particle coordinates before the collision step is required to ensure Galilean invariance. All particles are shifted by the *same* random vector with components in the interval $[-a/2, a/2]$ before the collision step. There is a great deal of freedom in how the rotation step is implemented, and any stochastic rotation matrix consistent with detailed balance can be used. In two dimensions, the stochastic rotation matrix is typically taken to be a rotation by an angle $\pm\alpha$, with probability 1/2 [1]. In three dimensions, two collision rules, denoted by models A and B in Ref. [11], have been considered. In model A [2], one performs rotations by an angle α about a randomly chosen axis. In model B [11], rotations are performed about one of three orthogonal rotation axes of a Cartesian coordinate system. At each collision step, one of these three axes is chosen at random, and a rotation by an angle $\pm\alpha$ is then performed, where the sign is chosen at random.

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 ν is given by [9,11]

$$\nu = \frac{\tau}{N k_B T} \sum_{n=0}^{\infty} \langle \sigma_{xy}(0) \sigma_{xy}(n\tau) \rangle, \quad (1)$$

where

$$\sigma_{xy}(n\tau) = -\frac{1}{\tau} \sum_j [v_{jx}(n\tau) \Delta \xi_{jy}(n\tau) + \Delta v_{jx}(n\tau) \Delta \xi_{jy}^s(n\tau)], \quad (2)$$

with $\Delta \xi_j(n\tau) = \xi_j((n+1)\tau) - \xi_j(n\tau)$, $\Delta \xi_j^s(n\tau) = \xi_j((n+1)\tau) - \xi_j^s((n+1)\tau)$, and $\Delta v_{xj}(n\tau) = v_{xj}((n+1)\tau) - v_{xj}(n\tau)$. $\xi_j(n\tau)$ is the cell coordinate of particle j at time $n\tau$, while ξ_j^s 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. [9] leads to three contributions to the transport coefficients, which were called the kinetic, rotational, and mixed terms. For a large mean free path, $\lambda = \tau \sqrt{k_B T} \gg a$, 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 ξ -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

$$\bar{\sigma}_{xy}^{kin}(n\tau) = -\sum_j v_{jx}(n\tau) v_{jy}(n\tau), \quad (3)$$

and

$$\bar{\sigma}_{xy}^{rot}(n\tau) = -\frac{1}{\tau} \sum_j B_{jy}(n\tau) v_{jx}(n\tau), \quad (4)$$

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

$$\langle B_{i\alpha}(n\tau) B_{j\beta}(m\tau) \rangle = \frac{a^2}{12} \delta_{\alpha\beta} (1 + \delta_{ij}) [2\delta_{n,m} - \delta_{n,m+1} - \delta_{n,m-1}], \quad (5)$$

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.

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

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

and

$$\nu_{rot} = \frac{\tau}{2N k_B T} \sum_{i,j=1}^N \{ \langle v_{ix}(0) v_{jx}(0) \rangle \langle B_{iy}(0) B_{jy}(0) \rangle + 2 \langle v_{ix}(0) v_{jx}(\tau) \rangle \langle B_{iy}(0) B_{jy}(\tau) \rangle \}. \quad (7)$$

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 [10], one finds

$$\nu_{kin}^{2D} = k_B T \tau \sum_{n=0}^{\infty} G_C(n\tau) = \frac{k_B T \tau}{2} \left[\frac{M}{(M-1 + e^{-M}) \sin^2(\alpha)} - 1 \right], \quad (8)$$

in two dimensions, where

$$G_C(n\tau) \equiv \langle \bar{\sigma}_{xy}^{kin}(0) \bar{\sigma}_{xy}^{kin}(n\tau) \rangle_C / N (k_B T)^2 = [1 - 2 \sin^2(\alpha) (M-1 + e^{-M}) / M]^n. \quad (9)$$

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

$$\nu_{kin}^{3D} = \frac{k_B T \tau}{2} \left[\frac{5M}{(M-1 + e^{-M}) [2 - \cos(\alpha) - \cos(2\alpha)]} - 1 \right] \quad (10)$$

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

Equations (8) and (10) are the same results one would obtain in the Chapman-Enskog approximation [1]. For a small mean free path, however, there are significant contributions to ν_{kin} that are neglected in this approximation. They arise from correlations between particles which are in the same (shifted) cell at more than one time step. Figure 1 contains a plot of the $G(n\tau) \equiv \langle \bar{\sigma}_{xy}^{kin}(0) \bar{\sigma}_{xy}^{kin}(n\tau) \rangle / N (k_B T)^2$, in two dimensions for $\alpha = 90^\circ$. The bullets are the result $G_C(n\tau)$ 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. (8) provides an excellent approximation for ν_{kin} . On the other hand, data obtained for $\lambda/a = 0.01$ (◦) exhibit much larger correlations for $n\tau \geq 2$. Figure 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 λ , we have

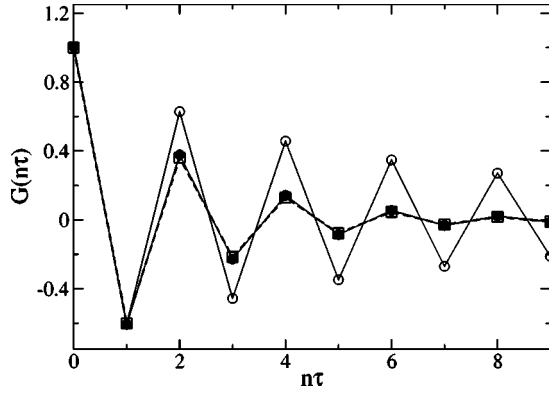


FIG. 1. $G(n\tau)$ as a function of time step for $\lambda/a=0.01$ (\circ) and $\lambda/a=1.0$ (\square). The bullets (\bullet) are the result $G_C(n\tau)$ given in Eq. (9). Averages were taken over 100 000 iterations and five different random number seeds. Parameters: $L/a=64$, $M=5$, and $\alpha=90^\circ$.

calculated $\delta G(2\tau)$ in the $\lambda \rightarrow 0$ 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 λ/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 [6,9,12]. The effect of these correlations on the value of the viscosity are less significant and only visible at intermediate mean free path, since they vanish for large λ 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. (7), 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 ν_{rot} in (7) obey the relation $\nu_{rot}^{diagonal} = -2\nu_{rot}^{off-diagonal}$. Using this result and relation (5), and averaging over the number of particles in a cell, one obtains [12]

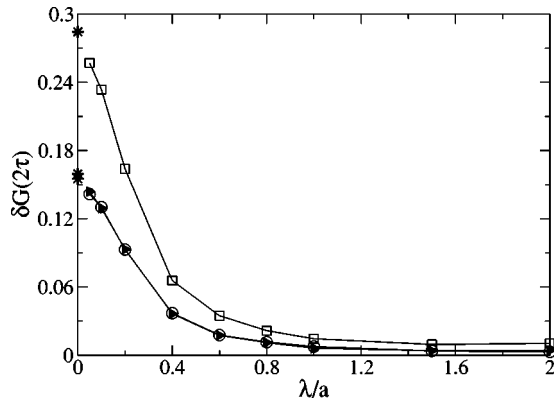


FIG. 2. $\delta G(2\tau)$ as a function of λ/a for rotation angles $\alpha=60^\circ$ (\circ), 90° (\square), and 120° (\blacktriangleright). The asterisks (*) are the theoretical values for $\lambda/a=0$. Time averages over 400 000 iterations for five different random number seeds were used to obtain the data. Parameters: $L/a=64$ and $M=5$.

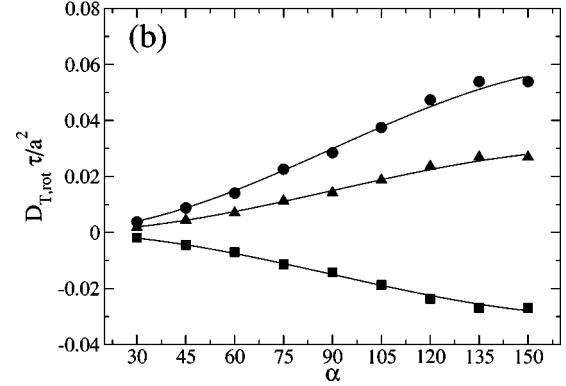
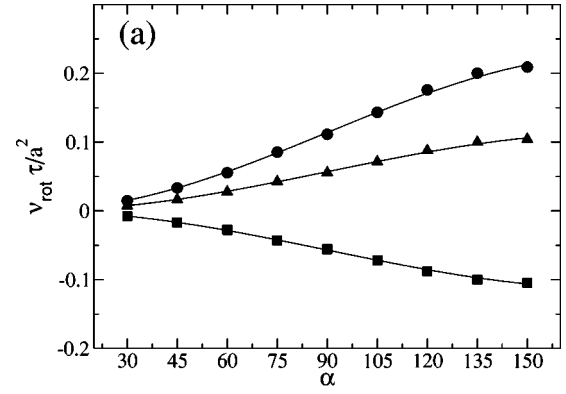


FIG. 3. Simulation results for the normalized rotational contribution to (a) the kinematic viscosity, $\nu_{rot}\tau/a^2$, and (b) the thermal diffusivity, $D_{T,rot}\tau/a^2$, as a function of the collision angle α . The circles (\bullet) are the diagonal, the squares (\blacksquare) the offdiagonal, and the triangles (\blacktriangle) 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 $\tau=1$.

$$\nu_{rot} = \frac{a^2}{6d\tau} \left(\frac{M-1+e^{-M}}{M} \right) [1 - \cos(\alpha)] \quad (11)$$

for all the collision models we considered [the standard model in $d=2$ and both models A and B in three dimensions (3D) [11]]. Equation (11) agrees with the result of Kikuchi *et al.* [10] obtained using a different nonequilibrium approach in shear flow, but deviates slightly for small M from the result given in Refs. [9,11]. Result (11) is compared with simulation data for the rotational contribution to the viscosity in Fig. 3(a).

The GK relation for the thermal diffusivity, D_T , derived in Refs. [8,9] can be resummed 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 two dimensions (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 $D_{T,rot}^{diagonal} = -2D_{T,rot}^{off-diagonal}$, which follows from energy conservation, can be used to show that the rotational contribution to the thermal diffusivity is

$$\begin{aligned}
D_{T,rot} &= \frac{a^2}{3(d+2)\tau M} \frac{1}{M} \left[1 - e^{-M} \left(1 + \int_0^M \frac{e^x - 1}{x} dx \right) \right] \\
&\quad \times [1 - \cos(\alpha)] \\
&\approx \frac{a^2}{3(d+2)\tau M} \frac{1}{M} \left(1 - \frac{1}{M} \right) [1 - \cos(\alpha)] \quad (12)
\end{aligned}$$

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 λ/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(b).

It is now clear that the random shift procedure introduced in Refs. [7,8] 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 nonequilibrium calculation of Kikuchi *et al.* [10], 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 nonequilibrium approach [13]. It can also be used to show that the bulk viscosity is equal to zero [9,12].

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