PHYSICAL REVIEW E **72**, 046707 (2005)

Equilibrium calculation of transport coefficients for a fluid-particle model

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(Received 19 May 2005; published 10 October 2005)

A recently introduced particle-based model for fluid flow, called stochastic rotation dynamics, can be made Galilean invariant by introducing a random shift of the computational grid before collisions. In this paper, it is shown how the Green-Kubo relations derived previously can be resummed to obtain exact expressions for the collisional contributions to the transport coefficients. It is also shown that the collisional contribution to the microscopic stress tensor is not symmetric, and that this leads to an additional viscosity. The resulting identification of the transport coefficients for the hydrodynamic modes is discussed in detail, and it is shown that this does not impose restrictions on the applicability of the model. The collisional contribution to the thermal conductivity, which becomes important for small mean free path and small average particle number per cell, is also derived.

DOI: 10.1103/PhysRevE.72.046707 PACS number(s): 47.11.+j, 05.40.−a, 02.70.Ns

I. INTRODUCTION

Particle-based simulation techniques have recently become an attractive alternative to more traditional methods for the coarse-grained modeling of a fluctuating solvent. A particularly appealing algorithm, introduced by Malevanets and Kapral [1,2], and later called multiparticle collision dynamics [3–6] or stochastic rotation dynamics (SRD) [7–14] provides a “hydrodynamic heat bath,” the details of which are not resolved, but which provides the correct hydrodynamic interactions and thermal fluctuations. The coarse-grained dynamics ignores molecular details, but enables simulations which span much longer time scales than can be addressed using molecular dynamics techniques. It has been used to study the behavior of polymers [5,15,16], colloids [2,6,17] (including sedimentation [14,18]), vesicles in shear flow [19], and complex fluids [20,21]. In addition to SRD’s numerical advantages, its simplicity has made 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 algorithms.

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, Galilean invariance can be restored by introducing a random shift [7,8] of the computational grid before every multiparticle interaction. In addition to restoring Galilean invariance, this grid shifting procedure accelerates momentum transfer between cells and leads to a collisional contribution to the transport coefficients. Two approaches have been used to analyze the resulting algorithm and determine the shear viscosity and thermal diffusivity. In Refs. [12,13], a nonequilibrium kinetic approach is used to derive the transport coefficients. In Refs. [7,8], a discrete-time projection operator technique was utilized to obtain Green-Kubo relations [22–24] for the model’s transport coefficients, and explicit expressions for the transport coefficients were derived in accompanying papers [9–11]. The two approaches are complementary and, for the most part, agree in their conclusions. The first is rather straightforward and intuitively appealing, but makes several assumptions which are not easily verified. The current approach justifies in detail several assumptions used in the nonequilibrium calculations of Refs. [12,13]; it can also be used to analyze the transport coefficients of the longitudinal modes, namely the bulk viscosity and thermal diffusivity, which are hard to calculate analytically in the nonequilibrium approach [13]. Note, in particular, that the collisional contribution to the thermal conductivity has not yet been determined using the nonequilibrium methods.

In this paper, we show in detail how the time series in the Green-Kubo relations for the transport coefficients can be resummed in such a way as to eliminate all dependence on the space-fixed cell coordinates of the particles [25]. This leads to a dramatic simplification of the sums, and enables an exact evaluation of the collisional contribution to the transport coefficients. It is also shown that there are only pure kinetic and collision contributions to the transport coefficients, as was implicitly assumed in the calculations of Refs. [12,13]. Explicit expressions for the collisional contributions to the viscosities and the thermal diffusivity are given, and the consequences of the fact that the collisional contribution to the microscopic stress tensor is *not* symmetric are discussed in detail. In particular, it is shown that this lack of symmetry leads to a slight change in the longitudinal viscous transport coefficient. The hydrodynamic modes are, however, not affected, and it does not impact on the applicability and stability of the method, even at low temperature.

II. SRD MODEL

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 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. Particles are then shifted back to their original positions after the collision. If we denote the cell coordinate of the shifted particle i by $\boldsymbol{\xi}_i^s$, the algorithm is summarized in the equations

$$\mathbf{r}_i(t+\tau)=\mathbf{r}_i(t)+\tau\mathbf{v}_i(t), \tag{1}$$

$$\mathbf{v}_i(t+\tau)=\mathbf{u}[\boldsymbol{\xi}_i^s(t+\tau)]+\boldsymbol{\omega}[\boldsymbol{\xi}_i^s(t+\tau)]\cdot\{\mathbf{v}_i(t)-\mathbf{u}[\boldsymbol{\xi}_i^s(t+\tau)]\}, \tag{2}$$

where $\boldsymbol{\omega}(\boldsymbol{\xi}_i^s)$ denotes a stochastic rotation matrix, and $\mathbf{u}(\boldsymbol{\xi}_i^s)\equiv 1/M\Sigma_{k\in\boldsymbol{\xi}^s}\mathbf{v}_k$ is the mean velocity of the particles in cell $\boldsymbol{\xi}^s$. All particles in the cell are subject to the same rotation, but the rotations in different cells are statistically independent. 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, $\boldsymbol{\omega}$, is typically taken to be a rotation by an angle $\pm\alpha$, with probability $1/2$ (see Refs. [7–9]). In three dimensions, two collision rules have been considered. In the first (model A in Ref. [10]), one performs rotations by an angle α about a randomly chosen direction, where all orientations of the random axis occur with equal probability. In the second (model B in Ref. [10]), rotations are performed about one of three orthogonal rotation axes, i.e., x-, y-, and z 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.

III. TRANSPORT COEFFICIENTS

Because of the cell structure introduced to define coarse-grained collisions, angular momentum is not conserved in a collision [26,27]. As a consequence, the macroscopic viscous stress tensor is not, in general, a symmetric function of the derivatives $V_{\alpha\beta}\equiv\partial_\alpha v_\beta$. Its general form can be determined as follows. Both the macroscopic viscous stress tensor, $\hat{\sigma}_{\alpha\beta}$, and the velocity gradient tensor, $V_{\alpha\beta}$ which appear in the Navier-Stokes equation, are rank two tensors. If the velocity gradients are small, we can assume—as is generally done—that the momentum transfer due to viscosity depends only on the first derivatives of the velocity, so that

$$\hat{\sigma}_{\alpha\beta}=C_{\alpha\beta\gamma\rho}V_{\gamma\rho}. \tag{3}$$

Symmetry arguments can be used to reduce the number of independent elements in the compliance tensor $C_{\alpha\beta\gamma\rho}$. Because of the simple cubic grid structure used in the algorithm, we have at least cubic symmetry. In this case, it can be shown [27] that the most general form for $C_{\alpha\beta\gamma\rho}$ is

$$C_{\alpha\beta\gamma\rho}/\rho=a\delta_{\alpha\beta}\delta_{\gamma\rho}+b\delta_{\alpha\gamma}\delta_{\beta\rho}+c\delta_{\alpha\rho}\delta_{\beta\gamma}+\epsilon\Gamma_{\alpha\beta\gamma\rho}, \tag{4}$$

where $\overset{\leftrightarrow}{\Gamma}$ is the rank four unit tensor [27], a, b, c, and ϵ are viscosity coefficients, and ρ is the mass density of the fluid. It follows that

$$\begin{aligned}\hat{\sigma}_{\alpha\beta}/\rho&=a\delta_{\alpha\beta}\partial_\lambda v_\lambda+b\partial_\alpha v_\beta+c\partial_\beta v_\alpha+\epsilon\Gamma_{\alpha\beta\gamma\rho}\partial_\gamma v_\rho\\&=\nu_1\left(\partial_\alpha v_\beta+\partial_\beta v_\alpha-\frac{2}{d}\delta_{\alpha\beta}\partial_\lambda v_\lambda\right)+\nu_2(\partial_\beta v_\alpha-\partial_\alpha v_\beta)\\&\quad+\gamma\delta_{\alpha\beta}\partial_\lambda v_\lambda+\epsilon\Gamma_{\alpha\beta\gamma\rho}\partial_\gamma v_\rho,\end{aligned} \tag{5}$$

with kinematic shear viscosities $\nu_1\equiv(b+c)/2$, $\nu_2\equiv(c-b)/2$, and bulk viscosity $\gamma=a+(b+c)/d$, where d is the spatial dimension. Here, ν_2 is the viscous transport coefficient associated with the nonsymmetric part of the stress tensor, and ϵ is a viscosity coefficient related to a possible lack of full rotational symmetry; both ν_2 and ϵ are usually zero in simple fluids. If we define a new tensor from the diagonal elements of $V_{\alpha\beta}$, namely $R_{\alpha\beta}\equiv\Gamma_{\alpha\beta\gamma\rho}V_{\gamma\rho}$, the resulting form of the momentum equation for a fluid reads

$$\rho\left[\frac{\partial\mathbf{v}}{\partial t}+(\mathbf{v}\cdot\boldsymbol{\nabla})\mathbf{v}\right]=-\boldsymbol{\nabla}p+\boldsymbol{\nabla}\cdot\overset{\leftrightarrow}{\hat{\sigma}} \tag{6}$$

$$\begin{aligned}&=-\boldsymbol{\nabla}p+\rho(\nu_1+\nu_2)\Delta\mathbf{v}\\&\quad+\rho\left(\left(1-\frac{2}{d}\right)\nu_1-\nu_2+\gamma\right)\boldsymbol{\nabla}(\boldsymbol{\nabla}\cdot\mathbf{v})\\&\quad+\epsilon\boldsymbol{\nabla}\cdot\overset{\leftrightarrow}{R},\end{aligned} \tag{7}$$

where p is the pressure. It can be seen from Eq. (7) that effective shear viscosity is $\nu=\nu_1+\nu_2$. In momentum space, the resulting linearized Navier-Stokes equation can be written as

$$\partial_t v_\alpha(\mathbf{k})=-\frac{\partial_\alpha p}{\rho}+\Lambda_{\alpha\beta}(\nu_1,\nu_2,\gamma,\epsilon;\hat{\mathbf{k}})v_\beta(\mathbf{k}), \tag{8}$$

where

$$\begin{aligned}\Lambda_{\alpha\beta}(\nu_1,\nu_2,\gamma,\epsilon;\hat{\mathbf{k}})\equiv{}&\nu_1\left(\delta_{\alpha\beta}+\frac{d-2}{d}\hat{k}_\alpha\hat{k}_\beta\right)+\nu_2(\delta_{\alpha\beta}-\hat{k}_\alpha\hat{k}_\beta)\\&+\gamma\hat{k}_\alpha\hat{k}_\beta+\epsilon\hat{k}_\gamma\hat{k}_\rho\Gamma_{\alpha\beta\gamma\rho}.\end{aligned} \tag{9}$$

$\Lambda_{\alpha\beta}(\nu_1,\nu_2,\gamma,\epsilon;\hat{\mathbf{k}})$ is the matrix of viscous transport coefficients. In a simple liquid, $\epsilon=0$ (because of invariance with respect to infinitesimal rotations), $\nu=\nu_1$, and $\nu_2=0$ (because the stress tensor is symmetric in $\partial_\alpha v_\beta$). In this case, Eq. (9) reduces to the well-known form [8]

$$\Lambda_{\alpha\beta}(\nu_1,\nu_2,\gamma,\epsilon;\hat{\mathbf{k}})=\nu\left(\delta_{\alpha\beta}+\frac{d-2}{d}\hat{k}_\alpha\hat{k}_\beta\right)+\gamma\hat{k}_\alpha\hat{k}_\beta. \tag{10}$$

As shown in Ref. [8], the discrete Green-Kubo (GK) relation

$$\Lambda_{\alpha\beta}(\nu_1,\nu_2,\gamma,\epsilon;\hat{\mathbf{k}}) \equiv \frac{\tau}{Nk_BT}{\sum_{n=0}^{\infty}}' \langle \hat{k}_\lambda \sigma_{\alpha\lambda}(0) | \hat{k}_{\lambda'} \sigma_{\beta\lambda'}(n\tau)\rangle, \tag{11}$$

for the SRD model can be used to express the matrix of viscous transport coefficients in terms of a sum of time correlation functions of the reduced fluxes $I_{1+\alpha}(\hat{\mathbf{k}},t)$

$$\hat{k}_\lambda \sigma_{\alpha\lambda}(t) \equiv I_{1+\alpha}(\hat{\mathbf{k}},t) = \frac{1}{\tau}\sum_j \Bigg(-[v_{j\alpha}(t)\hat{\mathbf{k}}\cdot\Delta\boldsymbol{\xi}_j(t) + \Delta v_{j\alpha}(t)\hat{\mathbf{k}}\cdot\Delta\boldsymbol{\xi}^s_j(t)] + \frac{\tau\hat{k}_\alpha}{d}v_j^2(t)\Bigg), \tag{12}$$

for $\alpha=1,\ldots,d$, with $\Delta\boldsymbol{\xi}_j(n\tau)=\boldsymbol{\xi}_j([n+1]\tau)-\boldsymbol{\xi}_j(n\tau)$, $\Delta\boldsymbol{\xi}^s_j(n\tau)=\boldsymbol{\xi}_j([n+1]\tau)-\boldsymbol{\xi}^s_j([n+1]\tau)$, and $\Delta v_{xj}(n\tau)=v_{xj}([n+1]\tau)-v_{xj}(n\tau)$. $\boldsymbol{\xi}_j(n\tau)$ is the cell coordinate of particle j at time $n\tau$, while $\boldsymbol{\xi}^s_j$ 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. (12) runs over all N particles of the system.

The corresponding expression for the thermal diffusivity is [8]

$$D_T = \frac{\tau}{c_pNk_BT^2}{\sum_{n=0}^{\infty}}' \langle \hat{k}_\lambda \chi_\lambda(0) | \hat{k}_{\lambda'}\chi_{\lambda'}(n\tau)\rangle, \tag{13}$$

with the energy flux

$$\chi_\lambda(n\tau) = \frac{1}{\tau}\sum_j \Bigg[(c_vT - v_j^2(n\tau)/2)\Delta\xi_{j\lambda}(n\tau) - \frac{1}{2}\Delta v_j^2(n\tau)\Delta\xi^s_{j\lambda}(n\tau) + \tau k_BTv_{j\lambda}(n\tau)\Bigg], \tag{14}$$

where $c_v=dk_B/2$ is the specific heat per particle at constant volume of an ideal gas and $\Delta v_j^2(n\tau)=v_j^2([n+1]\tau)-v_j^2(n\tau)$. The thermal conductivity, κ, is related to D_T by $\kappa=\rho c_pD_T$. Here and in the following, we have set the particle mass equal to 1.

IV. RESUMMED GREEN-KUBO RELATIONS

The straightforward evaluation of the GK relations [22–24] presented in Ref. [9] leads to three contributions to the transport coefficients, which were called the kinetic, rotational (or collisional), and mixed terms. The term "rotational" and the superscript "rot" refer to contributions from the collisions—stochastic rotations of the relative particle velocities—in the SRD model. For large mean-free path, $\lambda \gg a$, $\lambda=\tau\sqrt{k_BT}$, the assumption of molecular chaos is valid, and the kinetic contribution could be determined explicitly. For mean-free paths λ smaller than the cell size a, however, there were 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 $\Delta\xi$ in the stress correlation functions.

In fact, the appearance of $\Delta\xi$ is troubling, since one would not expect this 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. Consider first the time series ${\sum_{n=0}^{\infty}}'\hat{k}_\lambda\sigma_{\alpha\lambda}(n\tau)$. By canceling ξ-dependent terms in successive contributions to this series, it can be shown that

$${\sum_{n=0}^{\infty}}' \hat{k}_\lambda\sigma_{\alpha\lambda}(n\tau) = \frac{1}{2}\hat{k}_\lambda[A_{\alpha\lambda}(0)+A_{\alpha\lambda}(\tau)] + {\sum_{n=0}^{\infty}}'\hat{k}_\lambda\bar{\sigma}_{\alpha\lambda}(n\tau), \tag{15}$$

where $\bar{\sigma}_{\alpha\lambda}(n\tau)\equiv\bar{\sigma}^{kin}_{\alpha\lambda}(n\tau)+\bar{\sigma}^{rot}_{\alpha\lambda}(n\tau)$, with

$$\bar{\sigma}^{kin}_{\alpha\lambda}(n\tau) = -\sum_j [v_{j\alpha}(n\tau)v_{j\lambda}(n\tau) - \delta_{\alpha\lambda}v_j^2(t)/d],$$

$$\bar{\sigma}^{rot}_{\alpha\lambda}(n\tau) = -\frac{1}{\tau}\sum_j v_{j\alpha}(n\tau)B_{j\lambda}(n\tau), \tag{16}$$

$$A_{\alpha\lambda}(\tau) \equiv \frac{1}{\tau}\sum_j v_{j\alpha}(\tau)\Delta\xi^s_{j\lambda}(0), \tag{17}$$

and

$$B_{j\lambda}(n\tau) \equiv \xi^s_{j\lambda}([n+1]\tau) - \xi^s_{j\lambda}(n\tau) - \tau v_{j\lambda}(n\tau) = \Delta\xi_{j\lambda}(n\tau) - \Delta\xi^s_{j\lambda}(n\tau) + \Delta\xi^s_{j\lambda}([n-1]\tau) - \tau v_{j\lambda}(n\tau). \tag{18}$$

$B_{j\lambda}$ is a new stochastic variable which has very simple temporal correlations describing the geometrical properties of the underlying lattice.

Similarly, it can be shown that

$$\sigma_{\alpha\lambda}(0) = A_{\alpha\lambda}(0) - A_{\alpha\lambda}(\tau) + \bar{\sigma}_{\alpha\lambda}(0). \tag{19}$$

Using these results, the Green-Kubo relation (11) for the viscous transport coefficients can be written as the sum of two terms

$$\frac{\tau\hat{k}_\lambda\hat{k}_{\lambda'}}{Nk_BT}\Bigg\langle \frac{1}{2}[A_{\alpha\lambda}(0)A_{\beta\lambda'}(0) - A_{\alpha\lambda}(\tau)A_{\beta\lambda'}(\tau)] + \frac{1}{2}[A_{\alpha\lambda}(0)A_{\beta\lambda'}(\tau) - A_{\alpha\lambda}(\tau)A_{\beta\lambda'}(0)] + \frac{1}{2}[A_{\beta\lambda'}(0) + A_{\beta\lambda'}(\tau)]\bar{\sigma}_{\alpha bda}(0) + [A_{\alpha\lambda}(0) - A_{\alpha\lambda}(\tau)]{\sum_{n=0}^{\infty}}'\bar{\sigma}_{\beta\lambda'}(n\tau)\Bigg\rangle, \tag{20}$$

and

$$\frac{\tau}{Nk_BT}{\sum_{n=0}^{\infty}}'\langle\hat{k}_\lambda\bar{\sigma}_{\alpha\lambda}(0)|\hat{k}_{\lambda'}\bar{\sigma}_{\beta\lambda'}(n\tau)\rangle. \tag{21}$$

Stationarity implies that the first term in Eq. (20) equals zero, and that the last term reduces to

$$[A_{\alpha\lambda}(0) - A_{\alpha\lambda}(\tau)]\sum_{n=0}^{\infty}{}' \bar{\sigma}_{\beta\lambda'}(n\tau)$$

$$= -\frac{1}{2}[A_{\alpha\lambda}(\tau) + A_{\alpha\lambda}(0)]\bar{\sigma}_{\beta\lambda'}(0). \qquad (22)$$

Stationarity and time-reversal invariance imply that the remaining term also vanishes. Alternatively, the explicit form of $A_{\alpha\lambda}$, Eq. (17), can be used to show that this term vanishes. The expression in Eq. (20) is therefore zero, so that the Green-Kubo relation for the viscous transport coefficients is still given by (11), but with the stress tensor $\bar{\sigma}_{\alpha\lambda}$.

A similar calculation shows that the thermal diffusivity is given by (13), with $\chi_\lambda(n\tau)$ replaced by $\bar{\chi}_\lambda(n\tau) \equiv \bar{\chi}_\lambda^{kin}(n\tau) + \bar{\chi}_\lambda^{rot}(n\tau)$, with

$$\bar{\chi}_\lambda^{kin}(n\tau) = -\sum_j \left[-\frac{1}{2}v_j^2(n\tau)v_{j\lambda}(n\tau) + k_B T v_{j\lambda}(n\tau) \right],$$

$$\bar{\chi}_\lambda^{rot}(n\tau) = \frac{1}{2\tau}\sum_j v_j^2(n\tau)B_{j\lambda}(n\tau). \qquad (23)$$

Note that the new stress tensors do not depend on ξ, the space-fixed cell coordinates of the particles.

An alternative way to derive these results is to note that time-reversal invariance can be used to rewrite (11) and (13) as sums from $-\infty$ to $+\infty$. In this way, the discussion in the preceding paragraph of the $n=0$ term can be avoided.

V. CORRELATIONS INVOLVING B'S

$B_{i\alpha}(n\tau)$ is the α component of the difference between the change in the shifted cell coordinates during one streaming step and the actual distance traveled, $\tau v_{i\alpha}$. It has a number of important properties which simplify the calculation of the transport coefficients. In particular, it will be shown that all stress-stress correlation functions involving one **B** in the GK relations for the transport coefficients are zero, so that, for example, $\Lambda_{\alpha\beta}(\nu_1,\nu_2,\gamma,\epsilon;\hat{\mathbf{k}}) = \Lambda_{\alpha\beta}^{kin}(\nu_1,\nu_2,\gamma,\epsilon;\hat{\mathbf{k}}) + \Lambda_{\alpha\beta}^{rot}(\nu_1,\nu_2,\gamma,\epsilon;\hat{\mathbf{k}})$, with

$$\Lambda_{\alpha\beta}^{kin}(\nu_1,\nu_2,\gamma,\epsilon;\hat{\mathbf{k}}) \equiv \frac{\tau}{Nk_BT}\sum_{n=0}^{\infty}{}' \langle \hat{k}_\lambda \bar{\sigma}_{\alpha\lambda}^{kin}(0)|\hat{k}_{\lambda'}\bar{\sigma}_{\beta\lambda'}^{kin}(n\tau)\rangle, \qquad (24)$$

and

$$\Lambda_{\alpha\beta}^{rot}(\nu_1,\nu_2,\gamma,\epsilon;\hat{\mathbf{k}}) \equiv \frac{\tau}{Nk_BT}\sum_{n=0}^{\infty}{}' \langle \hat{k}_\lambda \bar{\sigma}_{\alpha\lambda}^{rot}(0)|\hat{k}_{\lambda'}\bar{\sigma}_{\beta\lambda'}^{rot}(n\tau)\rangle. \qquad (25)$$

The kinetic contributions to the viscosity were calculated previously in both two dimensions (2D) [9] and three dimensions (3D) [10], and will not be discussed here. Properties of the **B** correlations which enable an explicit evaluation of expression (25) are derived in the following subsections.

A. Factorization of $B-v$ correlations

The first one, $\langle B_{i\alpha}(n\tau)\rangle = 0$ for arbitrary n, implies that on average, the distance traveled by a particle during one time step is the average of difference of the shifted cell coordinates before and after the streaming step. This can be shown as follows. Consider

$$\langle B_{ix}(0)\rangle = \langle \xi_{ix}^s(\tau) - \xi_{ix}^s(0) - \tau v_{ix}(0)\rangle. \qquad (26)$$

The ensemble average includes averaging over the initial coordinates and velocities of all particles, as well as averages over the shift and collision matrix at each time step. Without loss of generality, assume that at $t=0$, the x coordinate of particle i is in the interval $[0,a)$. For $na \leq X \equiv x_i(0) + \tau v_{ix}(0) < (n+1)a$, the average of $\xi_{ix}^s(\tau) \equiv \xi_{ix}(\tau) - \Delta\xi_{ix}^s(0)$ over the random shift δ at time τ, denoted by $\langle\;\rangle_{\delta_\tau}$, at fixed particle coordinate and velocity, is [see Eq. (31) of Ref. [9]]

$$\langle \xi_{ix}^s(\tau)|_X\rangle_{\delta_\tau} = na - \langle \Delta\xi_{ix}^s|_X\rangle_{\delta_\tau} = X - a/2, \qquad (27)$$

so that

$$\langle B_{ix}(0)\rangle = \langle -a/2 + x_i(0) - \xi_{ix}^s(0)\rangle. \qquad (28)$$

Finally, averaging over the shift at $t=0$ gives $\langle \xi_{ix}^s(0)|_x\rangle_{\delta_0} = -a/2 + x_i(0)$, so that $\langle B_{ix}(0)\rangle = 0$.

Similar arguments can be used to show that the cross terms in the GK expressions for the transport coefficients involving one B are zero. For the shear viscosity, these terms involve correlations of the form $\langle v_{jx}(n\tau)v_{jy}(n\tau)v_{ix}(m\tau)B_{iy}(m\tau)\rangle$. Consider first the case $n=m=0$. Performing the average over the shift δ at $t=\tau$, the average reduces to

$$\langle v_{jx}(0)v_{jy}(0)v_{ix}(0)[y_i(0) - a/2 - \xi_{iy}^s(0)]\rangle. \qquad (29)$$

The average over the shift at $t=0$ yields zero because it does not affect the particle's initial velocities or positions. Consider now $m=0$ and $n=1$. In this case, first perform the average over the random shift at $t=0$. The result is

$$\langle v_{jx}(\tau)v_{jy}(\tau)v_{ix}(0)[\xi_{iy}^s(\tau) + a/2 - y_i(\tau)]\rangle. \qquad (30)$$

If the probability of any given configuration at $t=\tau$ in a shifted cell containing particle i is independent of δ, the average over the shift at $t=\tau$ factorizes. This is, in fact, the case since the average in (30) entails an integration over the initial particle coordinates and velocities at $t=0$. In this case, the average over δ can be performed; since $\langle \xi_{iy}^s(\tau) + a/2 - y_i(\tau)\rangle|_\delta$ vanishes, the result of this averaging is zero. An alternative, more detailed discussion of this proof is given in the Appendix.

The argument for general m and n is similar. Analogous reasoning can be used to show that correlations such as

$$\langle v_{ix}(0)v_{jx}(n\tau)B_{iy}(0)B_{jy}(n\tau)\rangle = \langle v_{ix}(0)v_{jx}(n\tau)\rangle\langle B_{iy}(0)B_{jy}(n\tau)\rangle \qquad (31)$$

factorize for arbitrary n.

B. Autocorrelation of B's

It is straightforward to evaluate equal-time correlation functions of the B variables. Using the results derived in the Appendix and Eq. (36) of Ref. [9], one has

$$\langle B_{ix}^2(0)\rangle = a^2/3 \text{ and } \langle B_{ix}(0)B_{jx}(0)\rangle = a^2/6, \quad \text{for } i \neq j. \tag{32}$$

Correlation functions such as $\langle B_{ix}(0)B_{ix}(\tau)\rangle$ can be evaluated as follows. Take $0 \leqslant x_i(0) < a$, $\Delta\xi_{ix} \equiv \Delta\xi_{ix}(0) = ma$, and $\Delta\xi_{ix}(\tau) = na$. Here and in the following expressions, the argument (0) will be omitted for clarity. Averaging over the random shift δ at $t = 2\tau$

$$\begin{aligned}\langle B_{ix}(0)B_{ix}(\tau)\rangle = &-\langle[(m+1/2)a - x_i(\tau)][\Delta\xi_{ix} + \Delta\xi_{ix}^s(-\tau) \\ &- \Delta\xi_{ix}^s - \tau v_{ix}]\rangle + \langle\Delta\xi_{ix}^s[\Delta\xi_{ix} + \Delta\xi_{ix}^s(-\tau) \\ &- \Delta\xi_{ix}^s - \tau v_{ix}]\rangle.\end{aligned} \tag{33}$$

Since $(m+1/2)a - x_i(\tau)$ does not depend on random shifts at time 0 and τ, the average over δ_0 and δ_τ in the first term on the right-hand side of Eq. (33) at fixed $x_i(0)$ and $v_{ix}(0)$ vanishes, so that

$$\langle B_{ix}(0)B_{ix}(\tau)\rangle = -\langle(\Delta\xi_{ix}^s)^2\rangle + \langle\Delta\xi_{ix}^s[\Delta\xi_{ix} + \Delta\xi_{ix}^s(-\tau) - \tau v_{ix}]\rangle. \tag{34}$$

Averaging the second term on the right-hand side of this equation over δ_0 and δ_τ, one finds

$$\begin{aligned}&\langle\Delta\xi_{ix}^s[\Delta\xi_{ix} + \Delta\xi_{ix}^s(-\tau) - \tau v_{ix}]\rangle \\ &= \frac{1}{a}\int_0^a dx_i \sum_{m=-\infty}^{\infty} \int_{(ma-x_i)/\tau}^{[(m+1)a-x_i]/\tau} [(m+1/2)a - x_i - \tau v_{ix}]^2 \\ &\times w(v_{ix})dv_{ix}.\end{aligned} \tag{35}$$

Comparing with Eqs. (18) and (32) of Ref. [9], it can be shown that (35) is equal to $-\langle(\Delta\xi_{ix})^2\rangle + 2\langle\Delta\xi_{ix}\Delta\xi_{ix}^s\rangle + \lambda^2 + a^2/12 = a^2/12$, where the last equality follows from

$$\langle\Delta\xi_{ix}\Delta\xi_{ix}^s\rangle = \frac{1}{2}[\langle\Delta\xi_{ix}^2\rangle - \lambda^2], \tag{36}$$

given as Eq. (36) in Ref. [9].

Finally, using (A3)

$$\langle B_{ix}(0)B_{ix}(\tau)\rangle = -a^2/6. \tag{37}$$

The average $\langle B_{ix}(0)B_{jx}(\tau)\rangle$ can be evaluated in a similar fashion. Take $m_0 a \leqslant x_i(0) < (m_0+1)a$, $n_0 a \leqslant x_j(0) < (n_0+1)a$, $\Delta\xi_{ix}(0) = m_1 a$, $\Delta\xi_{jx}(0) = n_1 a$, $\Delta\xi_{ix}(\tau) = m_2 a$ and $\Delta\xi_{jx}(\tau) = n_2 a$. Averaging over $\delta_{2\tau}$, one has

$$\begin{aligned}\langle B_{ix}(0)B_{jx}(\tau)\rangle = &-\langle[(n_0 + n_1 + 1/2)a - x_j(\tau)][\Delta\xi_{ix} + \Delta\xi_{ix}^s(-\tau) \\ &- \Delta\xi_{ix}^s - \tau v_{ix}]\rangle + \langle\Delta\xi_{jx}^s[\Delta\xi_{ix} + \Delta\xi_{ix}^s(-\tau) \\ &- \Delta\xi_{ix}^s - \tau v_{ix}]\rangle.\end{aligned} \tag{38}$$

Again, since $(n_0+n_1+1/2)a - x_i(\tau)$ does not depend on random shifts at time 0 and τ, the average over δ_0 and δ_τ in the first term on the right-hand side of Eq. (38) vanishes, so that

$$\begin{aligned}\langle B_{ix}(0)B_{jx}(\tau)\rangle = &-\langle\Delta\xi_{ix}^s\Delta\xi_{jx}^s\rangle + \langle\Delta\xi_{jx}^s[\Delta\xi_{ix} + \Delta\xi_{ix}^s(-\tau) \\ &- \tau v_{ix}]\rangle.\end{aligned} \tag{39}$$

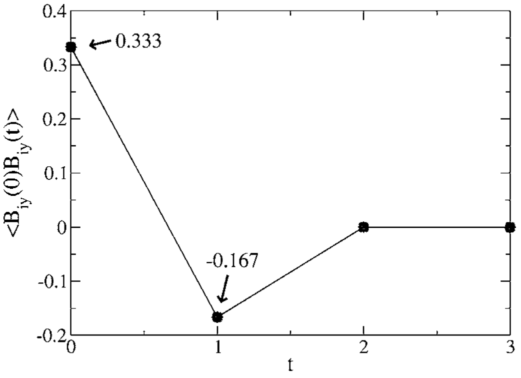

FIG. 1. $\langle B_{iy}(0)B_{iy}(t)\rangle$ as a function of time. Results for $\lambda/a = 0.05$, 0.10, 0.50, 1.00, and for collision angles $\alpha = 60°$, 90°, and 120° are plotted, indicating that there is no dependence on the value of the mean-free path. Time averages over 10^6 iterations were used to obtain the data. Parameters: $L/a = 32$ and $M = 5$.

Using Eqs. (B3), (C1), (C2), and (C4) from the Appendix, we have for $i \neq j$

$$\langle B_{ix}(0)B_{jx}(\tau)\rangle = -a^2/12. \tag{40}$$

All **B**-correlation functions for time lags greater than τ are zero. To understand this, consider $\langle B_{ix}(0)B_{jx}(2\tau)\rangle$. Averaging over $\delta_{3\tau}$

$$\begin{aligned}\langle B_{ix}(0)B_{jx}(2\tau)\rangle = &\langle[\Delta_{jx}(\tau) - (m+1/2)a + x_j(2\tau)] \\ &\times[\Delta\xi_{ix} + \Delta\xi_{ix}^s(-\tau) - \tau v_{ix}]\rangle,\end{aligned} \tag{41}$$

where ma is the cell coordinate of particle j at $t = 2\tau$. The second term in (41) has no dependence on $\delta_{2\tau}$, while average of the first term gives zero. Again, this requires that the probability of any given configuration in a shifted cell is independent of $\delta_{2\tau}$.

These results can be summarized by the relation

$$\langle B_{i\alpha}(n\tau)B_{j\beta}(m\tau)\rangle = \frac{a^2}{12}\delta_{\alpha\beta}(1 + \delta_{ij})[2\delta_{n,m} - \delta_{n,m+1} - \delta_{n,m-1}]. \tag{42}$$

Figure 1 presents simulation results for $\langle B_{iy}(0)B_{iy}(t)\rangle$ in $d = 2$ for various collision angles α and a range of mean-free paths. Figure 2 contains corresponding results for $\langle B_{iy}(0)B_{jy}(t)\rangle$. In both cases, the agreement with result (42) is excellent. Simulation results for $\langle v_{ix}(0)v_{ix}(t)B_{iy}(0)B_{iy}(t)\rangle$ as a function of time are presented in Fig. 3 for a similar range of parameters; the results are in agreement with the prediction of Sec. V A that this autocorrelation function factorizes, and that the resulting B correlations are given by Eq. (42).

It follows that there are only two—a pure kinetic and a pure rotational (or collision)—contributions to the transport coefficients. Relation (42) 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. Since the ki-

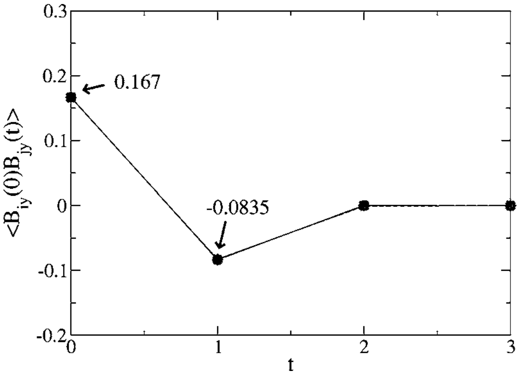

FIG. 2. $\langle B_{iy}(0)B_{jy}(t)\rangle$ as a function of time. Identical results are obtained for $\lambda=0.05$, 0.10, 0.50, 1.00 and for collision angles $\alpha=60°$, 90°, and 120°. Time averages over 10^6 iterations were used to obtain the data. Parameters: $L/a=32$ and $M=5$.

netic contribution to the stress tensor is symmetric and has been calculated elsewhere, we concentrate here on the (collisional) contributions arising from **B** correlations.

C. Viscosities

Explicit expressions for the collisional contributions to the viscous transport coefficients can be obtained by considering various choices for $\hat{\mathbf{k}}$ and α and β in Eq. (25) and using (9). Taking $\hat{\mathbf{k}}$ in the y direction and $\alpha=\beta=1$ yields

$$\nu^{rot} \equiv \nu_1^{rot} + \nu_2^{rot} = \frac{1}{\tau N k_B T} {\sum_{n=0}^{\infty}}' \sum_{i,j} \langle v_{ix}(0) B_{iy}(0) v_{ix}(t) B_{iy}(n\tau)\rangle . \tag{43}$$

Equation (43) is the expression used in Ref. [11] to determine the collisional contribution to the shear viscosity. ν_1^{rot} and ν_2^{rot} are the viscosities associated with the symmetric and the antisymmetric contributions to the matrix of viscous transport coefficients.

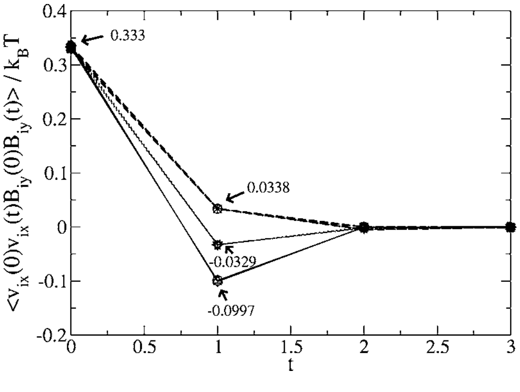

FIG. 3. $\langle v_{ix}(0)v_{ix}(t)B_{iy}(0)B_{iy}(t)\rangle$ as a function of time. Identical results are obtained for $\lambda=0.05$, 0.10, 0.50, and 1.00. Solid, dotted and dashed lines correspond to collision angles $\alpha=60°$, 90°, and 120°, respectively. Time averages over 10^6 iterations were used to obtain the data. Parameters: $L/a=32$ and $M=5$.

Other choices for $\hat{\mathbf{k}}$ and α and β yield expressions for other linear combinations of the transport coefficients. In particular, the choice $\hat{\mathbf{k}}=(1,0,0)$ and $\alpha=\beta=1$ yields

$$[1+(d-2)/d]\nu_1^{rot} + \gamma^{rot} + \epsilon^{rot} = \frac{1}{\tau N k_B T} {\sum_{n=0}^{\infty}}' \sum_{i,j} \langle v_{ix}(0) B_{ix}(0) v_{ix}(t) B_{ix}(n\tau)\rangle . \tag{44}$$

However, because of (42), the right-hand side of (44) is equal to (43), so that

$$[1+(d-2)/d]\nu_1^{rot} + \gamma^{rot} + \epsilon^{rot} = \nu^{rot}. \tag{45}$$

Finally, for $\hat{k}=(1,1,0)/\sqrt{2}$ and $\alpha=1$, $\beta=2$, one has

$$[(d-2)/d]\nu_1^{rot} - \nu_2^{rot} + \gamma^{rot} = 0, \tag{46}$$

since the resulting stress-stress correlation functions are zero. These results imply that $\epsilon^{rot}=0$, and that the longitudinal component of (9), which is the viscous contribution to the sound attenuation, is ν^{rot}. Finally, using these results in (9), it follows that the collision contribution to the macroscopic stress tensor is

$$\hat{\sigma}^{rot}_{\alpha\beta} = (\nu_1^{rot} + \nu_2^{rot})\partial_\beta v_\alpha = \nu^{rot}\partial_\beta v_\alpha, \tag{47}$$

up to a tensor $\overleftrightarrow{G}$ with vanishing divergence, $\partial_\beta G_{\alpha\beta}=0$, which therefore will not appear in the linearized hydrodynamic equations. The collisional contribution to the effective shear viscosity is therefore ν^{rot}, and the viscous contribution to the sound attenuation is also ν^{rot}, instead of the standard result, $2(d-1)\nu/d+\gamma$, for simple isotropic fluids. The corresponding hydrodynamic equation for the momentum density is therefore

$$\rho\left[\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v}\cdot\boldsymbol{\nabla})\mathbf{v}\right] = -\boldsymbol{\nabla} p + \rho(\nu^{kin}+\nu^{rot})\Delta\mathbf{v} + \frac{2-d}{d}\nu^{kin}\boldsymbol{\nabla}(\boldsymbol{\nabla}\cdot\mathbf{v}), \tag{48}$$

where we have used the fact that the kinetic contribution to the microscopic stress tensor, $\bar{\sigma}^{kin}$ in (16), is symmetric, and $\gamma^{kin}=0$ [9]. Note that there is no collisional contribution to the last term in Eq. (48). For $d=2$, the viscous contribution to the sound attenuation coefficient in a simple liquid is $\nu+\gamma$. The results of the current calculation are consistent with this result, with $\nu=\nu^{kin}+\nu^{rot}$ and $\gamma=0$ (as expected for a fluid with an ideal gas equation of state). In $d=3$, while the shear viscosity is still given by $\nu=\nu^{kin}+\nu^{rot}$, the viscous contribution to the sound attenuation coefficient is $4\nu^{kin}/3+\nu^{rot}$, instead of $4(\nu^{kin}+\nu^{rot})/3+\gamma$. The sound attenuation coefficient of the SRD model in three dimensions is therefore slightly smaller than in a simple liquid. Note, however, that the hydrodynamic equations are not affected. Although only certain linear combinations of the collisional contributions to the viscous transport coefficients are determined by Eqs. (43)–(46), all coefficients in the linearized hydrodynamic equations are uniquely determined.

Equilibrium measurements of the time-dependent density correlations [28] in two dimensions yield results for the sound attenuation coefficient which are in good agreement with the theoretical predictions. A detailed comparison of these simulation results with theory will be presented elsewhere [28]. Currently, there are no similar measurements in three dimensions.

The fact that the entropy of a fluid increases as a result of irreversible processes leads to certain positivity conditions on the transport coefficients [29]. SRD obeys an H-theorem [1,8], which implies that the entropy production is always non-negative. In Appendix E, it is shown, using a generalization of an argument from Ref. [29], that the requirement of a positive entropy production leads to the conditions $\nu^{kin}+\nu_1^{rot}\geqslant 0$, $\nu_2^{rot}\geqslant 0$, and $\gamma\geqslant 0$. Note that the result of Pooley and Yeomans [13] for the collisional stress tensor $\hat{\sigma}_{\alpha\beta}^{rot}=\nu^{rot}\partial_\beta v_\alpha$ amounts to assuming $\nu_1^{rot}=\nu_2^{rot}=\nu^{rot}/2$ and $\gamma=\nu^{rot}/d$.

VI. EXPLICIT EXPRESSIONS FOR THE COLLISIONAL CONTRIBUTIONS TO THE TRANSPORT COEFFICIENTS

A. Viscosities

Using the results of the previous sections, the collisional contribution to the viscosity can be written as

$$\nu^{rot}=\nu_1^{rot}+\nu_2^{rot}=\frac{1}{2\tau Nk_BT}\sum_{i,j=1}^{N}\{\langle v_{ix}(0)v_{jx}(0)\rangle\langle B_{iy}(0)B_{jy}(0)\rangle + 2\langle v_{ix}(0)v_{jx}(\tau)\rangle\langle B_{iy}(0)B_{jy}(\tau)\rangle\}. \tag{49}$$

It is straightforward to evaluate the various contributions to the right-hand side of (49). In particular, note that since velocity correlation functions only at equal time and for a time lag of one time step are required, molecular chaos can be assumed when evaluating these contributions, since it was shown in Ref. [11] that additional correlation effects only occur for larger time lags. Using (42), the first term on the right-hand side of Eq. (49) reduces to

$$\sum_{i,j=1}^{N}\langle B_{iy}(0)B_{jy}(0)\rangle\langle v_{ix}(0)v_{jx}(0)\rangle = \frac{a^2}{3}\sum_i\langle v_{ix}^2(0)\rangle+\frac{a^2}{6}\sum_i\sum_{j\neq i}\langle v_{ix}(0)v_{jx}(0)\rangle. \tag{50}$$

Momentum conservation, namely $\Sigma_k v_{kx}(0)=0$, can be used to write

$$\sum_i\sum_{j\neq i}\langle v_{ix}(0)v_{jx}(0)\rangle=-\sum_i\langle v_{ix}^2(0)\rangle, \tag{51}$$

so that the right-hand side of (50) reduces to

$$\frac{a^2}{6}\sum_i\langle v_{ix}^2(0)\rangle=\frac{a^2}{6}N_f k_BT, \tag{52}$$

where $N_f=N-1$, because of momentum conservation. Similarly, using momentum conservation at $t=\tau$, the second term in on the right-hand side of (49) reduces to

$$\sum_{i,j=1}^{N}\langle B_{iy}(0)B_{jy}(\tau)\rangle\langle v_{ix}(0)v_{jx}(\tau)\rangle=-\frac{a^2}{12}\sum_i\langle v_{ix}(0)v_{ix}(\tau)\rangle. \tag{53}$$

It follows that

$$\nu^{rot}=\frac{a^2}{12\tau}\left\{1-\frac{\langle v_{ix}(0)v_{ix}(\tau)\rangle}{k_BT}\right\}+O(1/N). \tag{54}$$

For $d=2$, if there are m_i particles in the collision cell $\xi_i^s(\tau)$, the ensemble average of the term in brackets in (54) is

$$1-\frac{1}{k_BT}\langle v_{ix}(0)v_{ix}(\tau)\rangle|_{m_i}=(1-1/m_i)[1-\cos(\alpha)], \tag{55}$$

where α is the collision angle. In three dimensions, the corresponding expression is

$$1-\frac{1}{k_BT}\langle v_{ix}(0)v_{ix}(\tau)\rangle|_{m_i}=\frac{2}{3}(1-1/m_i)[1-\cos(\alpha)], \tag{56}$$

for both models A and B. To obtain the final result, we now need to average over the number of particles, m_i, in the collision cell. If the average number of particles per cell is M, the probability that there are m_i particles in cell ξ_i^s is given by the Poisson distribution $P_p(m_i,M)=\mathrm{e}^{-M}M^{m_i}/m_i!$. The corresponding (normalized) probability that a given particle, i, is in a cell containing a total number of particles m_i is $m_iP_p(m_i,M)/M$. Averaging now over the number of particles in a cell, we have, finally

$$\nu^{rot}=\frac{a^2}{6d\tau}\left(\frac{M-1+e^{-M}}{M}\right)[1-\cos(\alpha)], \tag{57}$$

for all the collision models we considered (the standard model in $d=2$ and both models A and B in 3D). Equation (57) agrees with the result of Refs. [12,13] obtained using a completely different nonequilibrium approach in shear flow. Simulation results for the rotational contribution to the viscosity, Eq. (57), are compared with the theoretical prediction in Fig. 4 for small $M=3$, where our earlier approximation [7] for $M\gg 1$ would not be accurate. The new expression correctly describes the limit $M\to 0$, where the collisional viscosity should vanish.

B. Thermal diffusivity

The collisional contribution to the thermal diffusivity can be calculated in a similar fashion. In particular, taking $\hat{\mathbf{k}}=\hat{x}$ in (13) and (14), and using (23), we have $D_T=D_T^{kin}+D_T^{rot}$ with

$$D_T^{kin}=\frac{\tau}{c_pNk_BT^2}\sum_{n=0}^{\infty}{}'\sum_{i,j=1}^{N}\langle[v_i^2(0)/2-k_BT][v_j^2(n\tau)/2-k_BT]v_{ix}(0)v_{jx}(n\tau)\rangle, \tag{58}$$

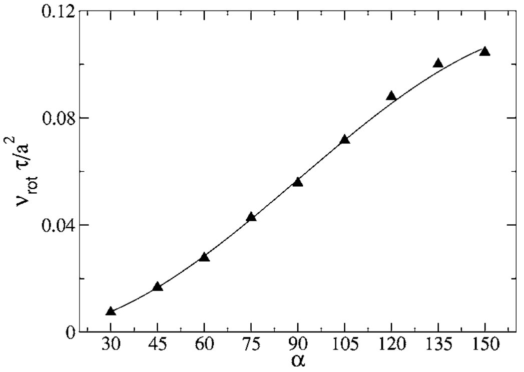

FIG. 4. The normalized collisional contribution to the kinematic viscosity, $\nu^{rot}\tau/a^2$, as a function of collision angle α. The solid line is the theoretical prediction (57). The data were obtained by time averaging over 360 000 iterations. Parameters: $L/a=16$, $\lambda/a=0.1$, $M=3$, and $\tau=1$.

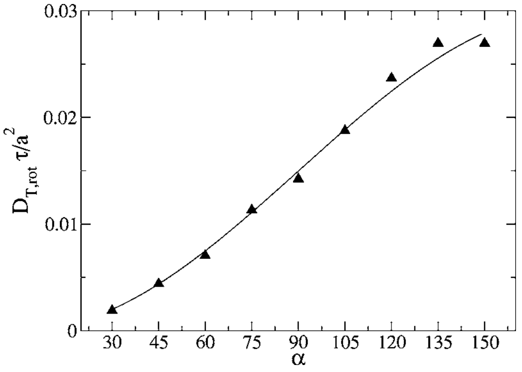

FIG. 5. The normalized collisional contribution to the thermal diffusivity, $D_T^{rot}\tau/a^2$, as a function of collision angle α. The solid line is the theoretical prediction (63). The data were obtained by time averaging over 360 000 iterations. Parameters: $L/a=16$, $\lambda/a=0.1$, $M=3$, and $\tau=1$.

$$D_T^{rot} = \frac{1}{8c_p\tau N k_B T^2}\sum_{i,j=1}^{N}\{\langle v_i^2(0)v_j^2(0)\rangle\langle B_{ix}(0)B_{jx}(0)\rangle + 2\langle v_i^2(0)v_j^2(\tau)\rangle\langle B_{ix}(0)B_{jx}(\tau)\rangle\}. \qquad (59)$$

The kinetic contributions to the thermal diffusivity were calculated previously in both 2D [9] and 3D [10]. Using the results presented earlier in this paper, it is straightforward to evaluate the collisional contribution to the thermal diffusivity. Just as momentum conservation was used to simplify the calculation of the collisional contribution to the viscosity, energy conservation, $\Sigma_{k=1}^{N}v_k^2=\text{const.}$, and (42) can be used to show that (59) reduces to

$$D_T^{rot} = \frac{a^2 d}{24\tau}\left\{1 - \frac{\langle v_i^2(0)v_i^2(\tau)\rangle}{d(d+2)(k_BT)^2}\right\}. \qquad (60)$$

For $d=2$, if there are m_i particles in the collision cell $\xi_i^s(\tau)$, the ensemble average of the term in brackets in (60) is

$$1 - \left.\frac{\langle v_i^2(0)v_i^2(\tau)\rangle}{8(k_BT)^2}\right|_{m_i} = \frac{1}{m_i}\left(1-\frac{1}{m_i}\right)[1-\cos(\alpha)]. \qquad (61)$$

In three dimensions, the corresponding expression is

$$1 - \left.\frac{\langle v_i^2(0)v_i^2(\tau)\rangle}{15(k_BT)^2}\right|_{m_i} = \frac{8}{15m_i}\left(1-\frac{1}{m_i}\right)[1-\cos(\alpha)], \qquad (62)$$

for both models A and B.

Using these results in (60) and averaging over the number of particles in a cell, assuming again that the probability of having a given particle, i, is in a cell containing a total number of particles m_i is $m_iP_p(m_i,M)/M$, where M is the average number of particles per cell, one finds

$$D_T^{rot} = \frac{a^2}{3(d+2)\tau M}\left[1 - e^{-M}\left(1+\int_0^M \frac{e^x-1}{x}dx\right)\right] \times[1-\cos(\alpha)], \qquad (63)$$

$$\xrightarrow[\text{large } M]{} \frac{a^2}{3(d+2)\tau M}\left(1+\mathrm{e}^{-M}(\ln M-1) - \frac{1}{M}-\frac{1}{M^2}-\frac{2}{M^3}-\cdots\right)[1-\cos(\alpha)], \qquad (64)$$

$$\xrightarrow[\text{small } M]{} \frac{a^2}{12(d+2)\tau}\left(M-\frac{5}{9}M^2+\cdots\right) \times[1-\cos(\alpha)], \qquad (65)$$

for all models considered. Note that in contrast to the viscosity, the rotational contribution to the thermal diffusivity is $O(1/M)$ for large M. Simulation results for the collisional contribution to thermal diffusivity are compared with (63) in Fig. 5. This contribution to the thermal diffusivity, which is not negligible for small M (such as $M=3$ in Fig. 5), was not discussed in Refs. [12,13].

A comparison of the relative size of the rotational and kinetic contributions to D_T is given in Fig. 6. Since D_T^{rot} is independent of temperature while D_T^{kin} increases linearly with temperature, there is a temperature (or mean-free path $\lambda=\tau\sqrt{k_BT}$) at which both contributions are equal. The ratio λ/a of this specific mean free path to the cell size a is plotted as a function of the rotation angle α. Results obtained using Eq. (63), Eq. (89) of Ref. [9], and Eq. (48) in Ref. [10] are presented for $M=5$ in both two (dashed line) and three dimensions (solid line). In two dimensions, D_T^{rot} can be larger than D_T^{kin} already at mean-free paths as large as $0.25a$ for large α. In $d=3$, the rotational contribution is slightly less important.

VII. CONCLUSION

It has been shown 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. The current approach justifies in detail several assumptions used in the nonequilibrium calculations of Refs. [12,13], and was used to

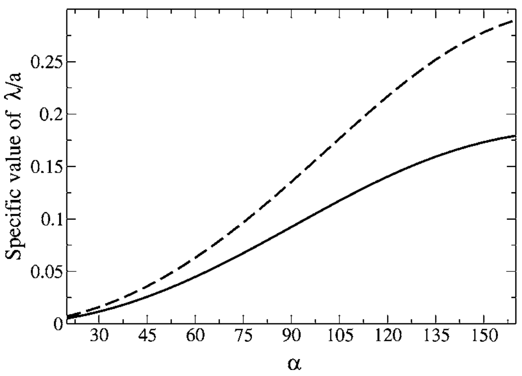

FIG. 6. The specific ratio, λ/a, at which $D_T^{kin}=D_T^{rot}$ as a function of the rotational angle α for $M=5$ in $d=2$ (dashed line) and $d=3$ (solid line). $\lambda=\tau\sqrt{k_BT}$ is the mean-free path, a is the cell size. For mean-free paths below the curve the rotational contribution to the thermal diffusivity, D_T^{rot}, is larger than the kinetic part, D_T^{kin}. Theoretical expressions given by Eq. (63), Eq. (89) of Ref. [9], and Eq. (48) of Ref. [10] were used.

determine the collisional contribution to the shear viscosity, the bulk viscosity, and the thermal diffusivity.

A detailed analysis of the consequences of the fact that SRD collisions do not conserve angular momentum was also presented. It was shown that, while the long-time, long-length-scale hydrodynamics of the model is not affected, it does lead to small changes in the viscous contribution to the sound attenuation coefficient. Although it has been pointed out previously [13] that the collisional contribution to the macroscopic viscous stress tensor is not symmetric, our interpretation of the consequences of this fact is different from that of Ref. [13]. In particular, the resulting slight modification of the coefficient of sound attenuation has no consequences for most practical applications, such as those in Refs. [16,19], and does not restrict the validity of the model.

ACKNOWLEDGMENTS

We thank J. Yeomans for helpful discussions which initiated this re-examination of the Green-Kubo approach. We also thank her and C.M. Pooley for making their unpublished notes available to us, and Alexander Wagner for numerous discussions. Support from the National Science Foundation under Grant Nos. DMR-0328468 and DMR-0513393, and ND EPSCoR through NSF Grant EPS-0132289, is gratefully acknowledged.

APPENDIX A: $\langle(\Delta\xi_{ix}^s)^2\rangle$

If $X_s=x_i+\delta$, with $0\leqslant x_i<a$

$$\langle\Delta\,\xi_{ix}^s|_X\rangle_\delta=\int_{-a/2}^{a/2}[\Theta(-X_s)-\Theta(X_s-a)]d\delta, \tag{A1}$$

and

$$\langle\,(\Delta\xi_{ix}^s)^2|_X\rangle_\delta=a\int_{-a/2}^{a/2}[\Theta(-X_s)+\Theta(X_s-a)]d\delta. \tag{A2}$$

Integrating over X, we have

$$\langle(\Delta\xi_{ix}^s)^2\rangle=\int_0^{a/2}d\delta\int_{a-\delta}^{a}dx_i+\int_{-a/2}^{0}d\delta\int_0^{-\delta}dx_i=a^2/4. \tag{A3}$$

APPENDIX B: $\langle\Delta\xi_{ix}^s\Delta\xi_{jx}^s\rangle$, $i\neq j$

$$\langle\Delta\xi_{ix}^s\Delta\xi_{jx}^s\rangle=\frac{1}{a}\int_{-a/2}^{a/2}d\delta\left\{\int_0^a dx_i[\Theta(-x_i-\delta)-\Theta(x_i+\delta-a)]\right\}^2. \tag{B1}$$

The integral over x_i is

$$\int_0^a dx_i[\Theta(-x_i-\delta)-\Theta(x_i+\delta-a)]=\delta\Theta(\delta)-\delta\Theta(-\delta), \tag{B2}$$

so that

$$\langle\Delta\xi_{ix}^s\Delta\xi_{jx}^s\rangle=\frac{1}{a}\int_{-a/2}^{a/2}d\delta[\delta^2\Theta(\delta)+\delta^2\Theta(-\delta)]=a^2/12. \tag{B3}$$

APPENDIX C: VARIOUS OTHER CORRELATIONS

There are a number of other useful relations which are required to evaluate the **B** correlations which can be easily evaluated using the same techniques. They include

$$\langle\Delta\xi_{ix}^s v_{ix}\rangle=\langle\Delta\xi_{ix}^s(-\tau)v_{ix}\rangle=0, \tag{C1}$$

and

$$\langle\Delta\xi_{ix}\Delta\xi_{jx}\rangle=\langle\Delta\xi_{ix}\Delta\xi_{jx}^s\rangle=0, \tag{C2}$$

$$\langle\Delta\xi_{ix}^s v_{jx}\rangle=\langle\Delta\xi_{ix}^s(-\tau)v_{jx}\rangle=0, \tag{C3}$$

and

$$\langle\Delta\xi_{ix}^s\Delta\xi_{jx}^s(-\tau)\rangle=\langle\Delta\xi_{ix}\Delta\xi_{jx}^s(-\tau)\rangle=0, \tag{C4}$$

for $i\neq j$. Finally

$$\langle\Delta\xi_{ix}^s(-\tau)[\Delta\xi_{ix}(0)-\Delta\xi_{ix}^s(0)]\rangle=-a^2/12. \tag{C5}$$

To show this, assume $0\leqslant x_i(0)<a$ and $na\leqslant x_i(0)+\tau v_{ix}(0)<(n+1)a$. Averaging first over δ at time τ and then the random shift at time $t=0$, for fixed $x_i(0)$ and $v_{ix}(0)$, one finds

$$\langle\Delta\xi_{ix}^s(-\tau)\,[\Delta\xi_{ix}(0)-\Delta\xi_{ix}^s(0)]|_{x_i(0),X}\rangle=\langle(a/2-x_i(0))(-a/2+x_i(0)-\tau v_{ix}(0))\rangle. \tag{C6}$$

The final ensemble average in (C6) reduces to

$$\frac{1}{a}\int_0^a dx\sum_{n=-\infty}^{\infty}(a/2-x)\int_{(na-x)/\tau}^{[(n+1)a-x]/\tau}dv_x[-a/2+x+\tau v_x]w(v_x)$$
$$=\frac{1}{a}\int_0^a dx(a/2-x)\int_{-\infty}^{\infty}dv_x[-a/2+x+\tau v_x]w(v_x), \tag{C7}$$

where we have dropped the index i and the time argument of

x and v_x for brevity. The integral over v_x can be performed immediately, and the remaining integral over x gives the result, $-a^2/12$.

APPENDIX D: PROOF OF RELATION $\langle v_{jx}(\tau)v_{jy}(\tau)v_{ix}(0)B_{iy}(0)\rangle=0$

In order to evaluate this expression, averages over the random shift at time $t=0$, $\delta_0\equiv(\delta_{0x},\delta_{0y})$, the shift at time $t=\tau$, $\delta_\tau\equiv(\delta_{\tau x},\delta_{\tau y})$, and over the initial positions, $\mathbf{r}_i(0)=[x_i(0),y_i(0)]$, and velocities, $\mathbf{v}_i(0)$ of *all* particles are required. Averaging first over δ_0, keeping all the other quantities fixed, yields Eq. (30). Next, note that $\xi^s_{iy}(\tau)$ has an implicit dependence on the initial positions and velocities at $t=0$ and δ_τ. We therefore write

$$\xi^s_{iy}(\tau)=\xi^s_{iy}(\tau;\delta_\tau,\{\mathbf{r}_k\},\{\mathbf{v}_k\}). \tag{D1}$$

Because of translational symmetry

$$\xi^s_{iy}(\tau;\delta_\tau,\{\mathbf{r}_k\},\{\mathbf{v}_k\})\equiv\xi^s_{iy}(\tau;0,\{\tilde{\mathbf{r}}_k\},\{\mathbf{v}_k\}), \tag{D2}$$

with $\tilde{\mathbf{r}}_k=\mathbf{r}_k+\delta_\tau$. Keeping $\{\tilde{\mathbf{r}}_k\}$ fixed, the average over δ_τ in (30) then become

$$\begin{aligned}
&\frac{1}{a}\prod_{k=1}^{N}\int_{-\infty}^{\infty}f(\{\mathbf{v}_k\})d\mathbf{v}_k\int_{-\infty}^{\infty}d\mathbf{r}_k\int_{-a/2}^{a/2}v_{jx}(\tau)v_{jy}(\tau)v_{ix}(0)[\xi^s_{iy}(\tau)\\
&\quad-y_i(0)+a/2-\tau v_{iy}(0)]d\delta_\tau\\
&=\frac{1}{a}\prod_{k=1}^{N}\int_{-\infty}^{\infty}f(\{\mathbf{v}_k\})d\mathbf{v}_k\int_{-\infty}^{\infty}d\tilde{\mathbf{r}}_k\int_{-a/2}^{a/2}v_{jx}(\tau)v_{jy}(\tau)v_{ix}(0)\\
&\quad\times[\xi^s_{iy}(\tau)-\tilde{y}_i(0)+\delta_\tau+a/2-\tau v_{iy}(0)]d\delta_\tau\\
&=\prod_{k=1}^{N}\int_{-\infty}^{\infty}f(\{\mathbf{v}_k\})d\mathbf{v}_k\int_{-\infty}^{\infty}d\tilde{\mathbf{r}}_k v_{jx}(\tau)v_{jy}(\tau)v_{ix}(0)[\xi^s_{iy}(\tau)\\
&\quad-\tilde{y}_i(0)+a/2-\tau v_{iy}(0)],
\end{aligned} \tag{D3}$$

where f is the N-particle Boltzmann distribution.

The remaining average over the initial configuration can be split up into a sum of several terms. Each term corresponds to a situation in which the particle labeled i is restricted at time zero to be in a specific cell ξ_1 together with k_1 other particles with given labels, while particle j is likewise residing only in a given cell ξ_2 together with a set of k_2 distinguishable particles. These restrictions are needed in order to have the postcollisional velocities $\mathbf{v}_j(\tau)$ and the cell label $\xi^s_{jy}(\tau)$ to be independent of the initial positions of the particles in every term.

We will show now that all these terms will vanish independently. Keeping the initial velocities fixed, an average over $\tilde{y}_i(0)$ is performed under the condition mentioned above, i.e., where $\xi^s_{jy}(\tau)=\xi_1$. These constraints mean that $\tilde{y}_i(0)$ must be in the interval $[\xi_1-\tau v_{iy}(0),\xi_1+a-\tau v_{iy}(0)]$. Since the postcollisional velocities of particle j are not affected by the position $\tilde{y}_i(0)$, each of the terms is proportional to

$$\begin{aligned}
&\int f(\{\mathbf{v}_i\})d\mathbf{v}_i^N\int_{\xi_1-v_{iy}(0)\tau}^{\xi_1+a-v_{iy}(0)\tau}\left[\xi_1+\frac{a}{2}-\tilde{y}_i(0)-\tau v_{iy}(0)\right]d\tilde{y}_i(0)\\
&\quad\propto a\left[\xi_1+\frac{a}{2}-\tau v_{iy}(0)\right]-\left.\frac{\tilde{y}_i(0)^2}{2}\right|_{\xi_1-\tau v_{iy}(0)}^{\xi_1+a-\tau v_{iy}(0)}=0.
\end{aligned} \tag{D4}$$

All terms vanish independently, so that $\langle v_{jx}(\tau)v_{jy}(\tau)v_{ix}(0)B_{iy}(0)\rangle$ is zero. Note that the same argument applies when $\xi_2=\xi_1$, i.e., the particles being in the same cell, for both $j\neq i$ and $j=i$.

APPENDIX E: ENTROPY PRODUCTION

The *general equation of heat transfer* is [29]

$$\rho T\left(\frac{\partial s}{\partial t}+\mathbf{v}\cdot\nabla s\right)=\hat{\sigma}_{\alpha\beta}\partial_\beta v_\alpha+\nabla\cdot(\kappa\nabla T), \tag{E1}$$

where the stress tensor $\hat{\sigma}_{\alpha\beta}$ is given in Eq. (5), s is the entropy per unit mass, ρ is the mass density, and κ is the thermal conductivity. It is easily verified that $\hat{\sigma}_{\alpha\beta}\partial_\beta v_\alpha$ can be written as

$$\begin{aligned}
\hat{\sigma}_{\alpha\beta}\partial_\beta v_\alpha=&\frac{\nu_1}{2}\left(\partial_\beta v_\alpha+\partial_\alpha v_\beta-\frac{2}{d}\delta_{\alpha\beta}\partial_\gamma v_\gamma\right)^2+\frac{\nu_2}{2}(\nabla\times\mathbf{v})^2\\
&+\gamma(\nabla\cdot\mathbf{v})^2,
\end{aligned} \tag{E2}$$

where all three terms are independent, invariant under coordinate transformations, and can vanish independently. Using (E1) and (E2), it can be shown [29] that the entropy production in a given volume Ω is

$$\begin{aligned}
\frac{d}{dt}\int_\Omega\rho s dV=&\int_\Omega\left[\frac{\kappa(\nabla T)^2}{T^2}+\frac{\nu_1}{2T}\left(\partial_\beta v_\alpha+\partial_\alpha v_\beta-\frac{2}{d}\delta_{\alpha\beta}\partial_\gamma v_\gamma\right)^2\right.\\
&\left.+\frac{\nu_2}{2T}(\nabla\times\mathbf{v})^2+\frac{\gamma}{T}(\nabla\cdot\mathbf{v})^2\right]dV.
\end{aligned} \tag{E3}$$

The requirement that the entropy production is non-negative implies that all transport coefficients in (E3) are greater than or equal to zero, namely

$$\nu_1\geqslant 0,\quad \nu_2\geqslant 0,\quad \gamma\geqslant 0,\quad \text{and } \kappa\geqslant 0. \tag{E4}$$

For $\nu_2=0$ this condition is identical to the well-known result discussed in Ref. [29]. For SRD, conditions (E4) reduce to

$$\nu^{kin}+\nu_1^{rot}\geqslant 0,\quad \nu_2^{rot}\geqslant 0,\quad \text{and } \gamma^{rot}\geqslant 0. \tag{E5}$$

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