Tagged particle in a sheared suspension: effective temperature determines density distribution in a slowly varying external potential beyond linear response

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Abstract. - We consider a sheared colloidal suspension under the influence of an external potential that varies slowly in space in the plane perpendicular to the flow and acts on one selected (tagged) particle of the suspension. Using a Chapman-Enskog type expansion we derive a steady state equation for the tagged particle density distribution. We show that for potentials varying along one direction only, the tagged particle distribution is the same as the equilibrium distribution with the temperature equal to the effective temperature obtained from the violation of the Einstein relation between the self-diffusion and tagged particle mobility coefficients. We thus prove the usefulness of this effective temperature for the description of the tagged particle behavior beyond the realm of linear response. We illustrate our theoretical predictions with Brownian dynamics computer simulations.

Introduction. – While the principles of equilibrium statistical mechanics are well established, a general framework of its non-equilibrium counterpart is still lacking. Recently, there has been a lot of interest in various facets of non-equilibrium statistical mechanics [1]. One of the recurring concepts is the so-called effective temperature [2]. Originally, it was defined through the violation of the fluctuation-dissipation relation in mean-field spin systems [3]. Subsequently, a variety of definitions of the effective temperature, in terms of violations of different dynamic and static linear response type relations, have been investigated using computer simulations of model fluids [4–7]. Notably, there have been few theoretical studies of the effective temperature [8–11].

The original definition of the effective temperature and almost all subsequent investigations mentioned above were concerned with the linear response regime. On the other hand, in equilibrium statistical mechanics the temperature plays an important role also outside the realm of linear response. Hayashi and Sasa [8] were the first to discuss an application of the effective temperature outside the linear response regime. They studied an exactly solvable model, a single Brownian particle in a tilted washboard potential. They showed that the effective temperature defined through the violation of the Einstein relation determines the density distribution in a long-wavelength external potential. Subsequently, there were three simulational studies of the effective temperature outside the linear response regime. Importantly, these studies dealt with strongly interacting many-body systems. Zamponi et al. [12] showed that the effective temperature enters into the fluctuation relation [13] for a driven glassy fluid. Ilg and Barrat [14] showed that the effective temperature determines the barrier crossing rate in a driven glassy system. Haxton and Liu [15] investigated the usefulness of the effective temperature for the description of material properties (specifically, the shear stress and the average inherent structure energy) of driven glassy systems. To date, there is no theoretical analysis of the effective temperature of a many-body system, outside the linear response regime.

Establishing the usefulness of the effective temperature outside the linear response regime is essential because, in principle, the violation of the fluctuation-dissipation relation does not have to be interpreted in terms of the effective temperature. The best illustration of this fact is the difference between two studies of the same exactly solvable model, a single Brownian particle in a tilted washboard potential. Hayashi and Sasa [8] used the violation of
the fluctuation-dissipation relation to define the effective
temperature. On the other hand, Speck and Seifert [10]
showed that the fluctuation-dissipation relation can be ef-
effectively restored if fluctuations of the Brownian particle
velocity are measured with respect to the local mean ve-
clocity. We should also mention a recent investigation [16]
which strives to provide a unifying framework for non-
equilibrium fluctuation-dissipation relations. It is similar
to Speck and Seifert’s approach in that it focuses on
an additive correction responsible for the violation of the
fluctuation-dissipation relation. It de-emphasizes the ef-
effective temperature altogether [17]. These examples show
an additive correction responsible for the violation of the
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temperatures defined in terms of the violation of the Ein-
stein relation along direction \( \hat{n} \). There
are two assumptions implicit in writing down these equa-
tions. First, hydrodynamic interactions are neglected.
Second, it is assumed that the shear rate is large enough
to drive Brownian particles out of equilibrium, but it is
small enough so that the solvent in which these particles
are suspended is in equilibrium. The latter assumption
implies that the Einstein relation for an isolated particle
is satisfied, \( D_0 = k_B T \mu_0 \), where \( T \) is the solvent

temperature.

In principle, in order to find the steady state tagged
particle distribution \( n_s \) we need to solve Eq. (1) and then
integrate over the positions of all other particles,

\[
n_s(\mathbf{r}_1) = \int d\mathbf{r}_2...d\mathbf{r}_N P_s^V(\mathbf{r}_1, ..., \mathbf{r}_N). \tag{3}
\]

In the absence of the shear flow, we know the solution of
Eq. (1) and we can easily find the equilibrium distribution
for the tagged particle in external potential \( V^\text{ext} \),

\[
n(\mathbf{r}_1) \propto \exp(-V^\text{ext}(\mathbf{r}_1)/(k_B T)). \tag{4}
\]

In the latter part of this Letter we use a Chapman-Enskog
type expansion to show that, in the presence of the shear
flow, for an external potential which varies slowly in the
plane perpendicular to the flow, the steady state tagged
particle distribution satisfies the following equation,

\[
\partial_{\mathbf{r}_\perp} \cdot (D \cdot \partial_{\mathbf{r}_\perp} + \mu \cdot (\partial_{\mathbf{r}_\perp} V^\text{ext}(\mathbf{r}_\perp))) n_s(\mathbf{r}_\perp) = 0. \tag{5}
\]

Here \( \mathbf{r}_\perp \) is a two-dimensional vector in the plane perpen-
dicular to the flow, and \( D \) and \( \mu \) are the self-diffusion and
the tagged particle mobility tensors which depend on the
suspension’s density, temperature, and shear rate.

It follows from Eq. (5) that, if external potential \( V^\text{ext} \)
varies only in the direction of unit vector \( \hat{n} \) (in the plane
perpendicular to the flow), the tagged particle distribution
has the form of the equilibrium distribution (4).

\[
n_s(\mathbf{r}_\perp \cdot \hat{n}) \propto \exp(-V^\text{ext}(\mathbf{r}_\perp \cdot \hat{n})/(k_B T^\text{eff}_\hat{n})), \tag{6}
\]

but with the temperature being replaced by the effective
temperature defined in terms of the violation of the Ein-
stein relation along direction \( \hat{n} \),

\[
T^\text{eff}_\hat{n} = \frac{\hat{n} \cdot D \cdot \hat{n}}{\hat{n} \cdot \mu \cdot \hat{n}}. \tag{7}
\]

For large enough shear rates the system can become struc-
turally anisotropic even though the density is still uniform.
In particular, structure factors along different directions

\[\text{Tagged particle density distribution.} \quad \text{– The } N-
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equation,

\[
\left[ \Omega_s + \partial_{\mathbf{r}_1} \cdot \left( D_0 \partial_{\mathbf{r}_1} - \mu_0 \sum_{i \neq j=1}^N F(\mathbf{r}_{ij}) - \mathbf{v}(\mathbf{r}_i) \right) \right] P_s^V(\mathbf{r}_1, ..., \mathbf{r}_N) = 0, \tag{1}
\]

where \( \Omega_s \) is the Smoluchowski operator with shear flow,

\[
\Omega_s = \sum_{i=1}^N \partial_{\mathbf{r}_i} \cdot \left( D_0 \partial_{\mathbf{r}_i} - \mu_0 \sum_{i \neq j=1}^N F(\mathbf{r}_{ij}) - \mathbf{v}(\mathbf{r}_i) \right). \tag{2}
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In the above equations, \( \mu_0 \) and \( D_0 \) is the mobility and the
diffusion coefficient, respectively, of an isolated Brownian
particle, \( F(\mathbf{r}_{ij}) \) is the force acting on particle \( i \) due to
particle \( j \), \( \mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j \), and \( \mathbf{v} \) is the uniform shear flow,
\( \mathbf{v}(\mathbf{r}_i) = \Gamma \cdot \mathbf{r}_i \), with \( \mathbf{\Gamma} \) being the velocity gradient tensor.
Finally, \( V^\text{ext} \) is an external potential. It acts only on the
tagged particle and varies in the plane perpendicular to the
flow, \( \partial_{\mathbf{r}_1} \cdot \Gamma V^\text{ext}(\mathbf{r}_1) = 0 \).

We should emphasize that Eqs. (1-2) without the exter-
nal potential have been widely used in investigations of the
dynamics and rheology of sheared suspensions [19]. There
are two assumptions implicit in writing down these equa-
tions. First, hydrodynamic interactions are neglected.
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particle, \( F(\mathbf{r}_{ij}) \) is the force acting on particle \( i \) due to

in the reciprocal space vary differently with the shear rate \[7\]. In principle, one cannot expect that in such cases the effective temperature is isotropic. On the other hand, for a system with very slow dynamics (e.g., a glassy system) there is a range of shear rates for which the system is strongly out of equilibrium but still structurally isotropic, with essentially shear-rate-independent structure factors along all directions in the reciprocal space \[4\]. In such cases it has been found that the effective temperature is isotropic in the linear response regime \[4\]. We expect (although we did not investigate it) that the same will be true also beyond the realm of the linear response. Of course, for any ergodic system \( T_{\text{eff}} \to T \) in the limit \( \gamma \to 0 \).

One of the consequences of Eq. \( 6 \) is the following relation between an infinitesimally small external potential \( \delta V_{\text{ext}} \) and the resulting change \( \delta n_s \) of the tagged particle distribution,

\[
\delta n_s(r_\perp \cdot \hat{n}) = -\frac{n_s}{k_B T_{\text{eff}}^s} \delta V_{\text{ext}}(r_\perp \cdot \hat{n}).
\]

Eq. \( 5 \) is a generalization of the familiar equilibrium linear response relation. It is analogous to static linear response type relations that were used to define effective temperatures by A. Liu and collaborators \[5\]. Importantly, in Eq. \( 8 \) effective temperature \( T_{\text{eff}} \) appears in the place of a static linear response-based effective temperature. This is the first time that the effective temperature defined in terms of the violation of a dynamic fluctuation-dissipation relation has been shown theoretically to be identical to the effective temperature defined in terms of the violation of a static linear response relation.

**Brownian dynamics simulations.** — We performed Brownian dynamics computer simulations as described in Ref. \[7\]. The system consists of \( N = 1372 \) particles interacting via a screened Coulomb potential,

\[
V(r) = A \exp\left(-\kappa(r - \sigma)\right)/r,
\]

with \( A = 475 k_B T \sigma \) and \( \kappa \sigma = 24 \). We simulated the system at a dimensionless density, \( N \sigma^3/L^3 \), equal to 0.408 (\( L \) is the box length), which corresponds to the hard-sphere volume fraction equal to 0.43 \[20\]. We applied shear flow in the \( x \) direction with the velocity gradient in the \( y \) direction, \( \Gamma = \gamma \hat{x} \hat{y} \) with \( \gamma \) being the shear rate and \( \hat{\alpha} \) the unit vector along the \( \alpha \) axis. In the following we use reduced units: we measure lengths in units of \( \sigma \), time in units of \( \sigma^2/D_0 \), energy in units of \( k_B T \), and effective temperature in units of \( T \).

To illustrate the validity of Eq. \( 6 \) we introduced a slowly varying external potential acting on particle number 1 \[21\]. We chose the simplest, plane wave potential,

\[
V_{\text{ext}}(\alpha_1) = V_0 \sin(2\pi \alpha_1 / L),
\]

where \( \alpha = y, z \). Note that \( 2\pi / L \) is the smallest wave-vector allowed by the periodic boundary conditions. We used \( V_0 = 2 \), which is well outside the linear response regime (see Fig. \( 1 \)). For each choice of the potential we performed 4 independent runs at the following shear rates: 0, 5, 10, and 20. We monitored the tagged particle density distribution, \( n_s(\alpha), \alpha = y, z \). All results presented are averaged over 4 runs.

In Fig. 1 we show tagged particle steady state density distributions. Qualitatively, with increasing shear rate the influence of the external potential decreases, which could be interpreted as an increasing effective temperature. More importantly, the simulations’ results can be fitted very well using equilibrium-like distributions \( 6 \) with an effective temperature being the only fit parameter. In Fig. 2 we show that the values of \( T_{\text{eff}}^\alpha, \hat{\alpha} = y, z \), obtained from the fits agree very well with the previously obtained effective temperatures defined through the violation of the Einstein relation \[7\].

At the largest shear rates we found previously that the system is structurally anisotropic \[7\]. Thus, in principle, different effective temperatures can be obtained along different directions. We note that results showed in Fig. 2 suggest slightly different effective temperatures along \( \hat{y} \) and \( \hat{z} \) directions. However, the difference between these temperatures is within error bars of our simulations.

**Derivation.** — We make two assumptions about the external potential acting on the tagged particle. First, we assume that it is transverse, i.e., that it varies only in the plane perpendicular to the flow, \( \partial_{\mathbf{r}_1} \cdot \mathbf{V}_{\text{ext}}(\mathbf{r}_1) = 0 \). Second, we assume that the potential is slowly varying. To make the latter assumption explicit and to facilitate the subsequent gradient expansion we introduce a small parameter \( \epsilon \) and write the external potential as \( V_{\text{ext}}(\mathbf{r}_1) \). As usual, at the end of the calculation we will put \( \epsilon = 1 \). We emphasize that no assumption is made regarding the strength of the external potential.

It is natural to assume that for a slowly varying, trans-
verse external potential the tagged particle density distribution \(n_s\) will also be slowly varying and transverse. To find a steady state equation for \(n_s\) we use the Chapman-Enskog \([22]\) expansion. Specifically, we follow Titulaer \([23]\) who applied the Chapman-Enskog expansion to the problem of deriving the so-called contracted, Smoluchowski description of a Brownian particle in position space from the full Fokker-Planck description in velocity and position space. We note that for linear equations considered in Ref. \([23]\) and in our Letter, the Chapman-Enskog procedure is closely related to a variant of perturbation theory \([24]\).

Before applying the Chapman-Enskog expansion, we have to face the fact that, although the tagged particle density \(n_s\) is slowly varying, due to the inter-particle interactions \(N\)-particle distribution \(P_s^V\) is not a slowly varying function of the tagged particle position \(r_1\). To disentangle the slow variation of \(P_s^V\) with \(r_1\), which is induced by the external potential, and the rapid variation \(P_s^V^r\) with \(r_1\), which originates from the inter-particle interactions, we introduce new variables: \(R_1 = cr_1, R_2 = r_{21},…, R_N = r_{N1}\). In these new variables the steady state equation \((11)\) has the following form:

\[
\left[\Omega^{(0)} + \epsilon \Omega^{(1)} + \epsilon^2 \Omega^{(2)}\right] P_s^V(R_1,…,R_N) = 0. \tag{11}
\]

In Eq. \((11)\),

\[
\begin{align*}
\Omega^{(0)} &= -\partial_{R_1} \cdot \Gamma \cdot R_1 + \sum_{i=2}^{N} \partial_{R_i} \cdot \left( D_0 \partial_{R_i} + D_0 \sum_{j=2}^{N} \partial_{R_j} \right) \\
&\quad - \mu_0 \sum_{i \neq j=2}^{N} F(R_{ij}) + \mu_0 \sum_{j=2}^{N} F(-R_j) - \partial_{R_1} \cdot \Gamma \cdot R_1 \\
\Omega^{(1)} &= -\partial_{R_1} \cdot \left( D_0 \sum_{i=2}^{N} \partial_{R_i} + \mu_0 \sum_{i=2}^{N} F(-R_i) \right) \\
&\quad - \sum_{i=2}^{N} \partial_{R_i} \cdot \mu_0 \left( \partial_{R_i} V^\text{ext}(R_1) \right) - D_0 \sum_{i=2}^{N} \partial_{R_i} \cdot \partial_{R_i} V^\text{ext}(R_1) \\
\Omega^{(2)} &= \partial_{R_1} \cdot \left( D_0 \partial_{R_1} + (\partial_{R_1} V^\text{ext}(R_1)) \right).
\end{align*}
\tag{12}
\]

We now follow Titulaer \([23]\) and simultaneously find a special solution of Eq. \((11)\)

\[
P_s^V(R_1,…,R_N) = n_s(R_1)P_s^{(0)}(R_2,…,R_N) + \epsilon P_s^{(1)}(R_1,R_2,…,R_N) + \epsilon^2 P_s^{(2)}(R_1,R_2,…,R_N) + \cdots
\]

and a steady state equation for the tagged particle distribution,

\[
\left( D^{(0)} + \epsilon D^{(1)} + \epsilon^2 D^{(2)} + \cdots \right) n_s(R_1) = 0. \tag{16}
\]

Here Eq. \((16)\) is obtained from integrating Eq. \((11)\) over \(R_i, i > 1\). Thus, e.g.,

\[
D^{(0)} n_s(R_1) = \int dR_2…dR_N \Omega^{(0)} n_s(R_1)P_s^{(0)}(R_2,…,R_N), \tag{17}
\]

and

\[
D^{(1)} n_s(R_1) = \int dR_2…dR_N \Omega^{(1)} n_s(R_1)P_s^{(0)}(R_2,…,R_N) + \int dR_2…dR_N \Omega^{(0)} P_s^{(1)}(R_1,R_2,…,R_N). \tag{18}
\]

It should be emphasized that in the Chapman-Enskog type procedure the steady state tagged particle distribution \(n_s\) is not expanded in \(\epsilon\). Moreover, higher order terms \(P_s^{(i)}, i > 0\), are determined up to an arbitrary multiple of \(P_s^{(0)}\). \([23]\). We follow the usual procedure \([23]\) and impose conditions

\[
\int dR_2…dR_N P_s^{(i)}(R_1,…,R_N) = 0 \text{ for } i > 0,
\]

which fix these contributions to be zero. It should be noted that due to these conditions the tagged particle distribution \(n_s\) is completely determined by the first term in expansion \((15)\).

We substitute \((15)\) into steady state equation \((11)\), solve iteratively for \(P_s^{(i)}\), and calculate \(D^{(i)}\). The results of this procedure are as follows. First, \(P_s^{(0)}\) is the translationally invariant solution of the Smoluchowski equation with shear flow in the absence of an external potential. This solution depends only on \(R_1, i > 1\), and is normalized in the following way: \(\int dR_1...dR_N P_s^{(0)} = 1\). Second, \(D^{(0)} = \partial_{R_1} \cdot \Gamma \cdot R_1\). Thus, for \(n_s(R_1)\) that varies only in the plane perpendicular to the flow \(D^{(0)}\) does not contribute to steady state equation \((15)\). Third, \(D^{(1)}\) vanishes. Fourth, \(D^{(2)}\) is determined by the following equation

\[
\begin{align*}
D^{(2)} n_s(R_1) &= \left[ D_0 \partial_{R_1} \cdot \partial_{R_1} + \partial_{R_1} \cdot \left( \partial_{R_1} V^\text{ext}(R_1) \right) \right] n_s(R_1) \\
&\quad - \partial_{R_1} \cdot \int dR_2…dR_N \mu_0 \sum_{i=2}^{N} F(-R_i) P_s^{(1)}(R_1,…,R_N).
\end{align*}
\tag{20}
\]
To complete the calculation, we determine $P_s^{(1)}$ using terms of order $\epsilon$ in Eq. (11),

\[ \Omega^{(0)} P_s^{(1)}(R_1, ..., R_N) = -\Omega^{(1)} n_s(R_1) P_s^{(0)}(R_2, ..., R_N). \]  \hspace{1cm} (21)

Using the explicit form of $\Omega^{(1)}$, Eq. (13), it can be showed that the source at the right-hand-side of Eq. (21) consists of two terms, which are proportional to $[\partial_{R_i} n_s(R_1)]$ and $[\partial_{R_i} V^{ext}(R_1)] n_s(R_1)$:

\[ -\Omega^{(1)} n_s(R_1) P_s^{(0)}(R_2, ..., R_N) = \left[ \partial_{R_i} n_s(R_1) \right] \left( 2D_0 \sum_{i=2}^{N} \partial_{R_i} + \mu_0 \sum_{j \neq 1} F(-R_j) \right) \times P_s^{(0)}(R_2, ..., R_N) \]

\[ + [\partial_{R_i} V^{ext}(R_1)] n_s(R_1) \mu_0 \sum_{i=2}^{N} \partial_{R_i} P_s^{(0)}(R_2, ..., R_N). \]  \hspace{1cm} (22)

To get $P_s^{(1)}$ we now have to solve Eq. (21). While inverting operator $\Omega^{(0)}$, we note that since both $n_s(R_1)$ and $V^{ext}(R_1)$ vary only in the plane perpendicular to the flow, operator $[\Omega^{(0)}]^{-1}$ will not act on them. Therefore, $P_s^{(1)}$ also consists of two terms which are proportional to $[\partial_{R_i} n_s(R_1)]$ and $[\partial_{R_i} V^{ext}(R_1)] n_s(R_1)$. After these two terms are substituted into Eq. (20) we can easily derive the following equality

\[ D^{(2)} n(R_1) = [\partial_{R_i} \cdot D \cdot \partial_{R_i} + \partial_{R_i} \cdot \mu \cdot (\partial_{R_i} V^{ext}(R_1))] n(R_1). \] \hspace{1cm} (23)

We now show that tensors $D$ and $\mu$, which enter into Eq. (23), are the self-diffusion tensor and the tagged particle mobility tensor, respectively. Explicitly,

\[ D = D_0 I + D^{int} \] \hspace{1cm} (24)

where $I$ is the unit tensor and the interaction contribution $D^{int}$ is given by the following formula,

\[ D^{int} = \frac{1}{V} \int dr_1 ... dr_N N \sum_{i=2}^{N} \int \frac{\partial_{R_i} F(-R_i)}{[\Omega^{(0)}]^{-1}} \times \left( -2D_0 \sum_{i=2}^{N} \partial_{R_i} + \mu_0 \sum_{j \neq 1} F(-R_j) \right) \times P_s^{(0)}(R_2, ..., R_N). \] \hspace{1cm} (25)

After returning to the original variables $r_1, ..., r_N$ and putting $\epsilon = 1$, the interaction contribution $D^{int}$ can be written in the following form:

\[ D^{int} = \frac{1}{V} \int dr_1 ... dr_N \mu_0 F_1 \Omega_s^{-1} \left( 2D_0 \partial_{r_1} - \mu_0 F_1 \right) \times P_s^{(0)}(r_1, ..., r_N). \] \hspace{1cm} (26)

where $F_1$ is the force acting on the tagged particle, $F_1 = \sum_{i=2}^{N} F(r_{1i})$. Following very similar steps one can show that

\[ \mu = \mu_0 I + \mu^{int} \] \hspace{1cm} (27)

where $\mu^{int} = \frac{1}{V} \int dr_1 ... dr_N \mu_0 F_1 \Omega_s^{-1} D_0 \partial_{r_1} P_s^{(0)}(r_1, ..., r_N).$ \hspace{1cm} (28)

The above expressions are identical to the expressions for the self-diffusion tensor and the tagged particle mobility tensor that have been derived in Refs. [9, 11].

Finally, we return to the original variables in Eq. (23), put $\epsilon = 1$, use the fact that both the external potential and the tagged particle density vary only in the plane perpendicular to the flow, and arrive at Eq. (5).

Discussion. – We showed that the effective temperature defined through the violation of the Einstein relation determines the tagged particle density distribution induced by a slowly varying external potential acting on the tagged particle. As a consequence, the same effective temperature enters into the violation of the Einstein relation and the modified static linear response relation for the tagged particle. It would be very interesting to extend both results to at least some collective properties. This would provide additional fundamental justification for introducing the effective temperature. It would also allow some deeper theoretical understanding of the very intriguing observation made by Liu and collaborators [5] that for some combinations of variables one should use dynamic linear response relations to determine the effective temperature whereas for other combinations of variables one could use static linear response relations.

We note in this context that the most obvious (and perhaps the most naive) generalization of the present results to the collective density is not valid already in the linear response regime: we showed earlier [7] that the effective temperature obtained from the collective version of the modified static response relation does not agree with the effective temperature defined through the violation of the Einstein relation.

It would also be very interesting to extend the present results, which pertain to non-linear response to an external potential, to material properties, in the spirit of the numerical investigation of Haxton and Liu [15].

The present results (and essentially all previous work on fluctuation-dissipation relation violation in systems of interacting particles) are concerned with a fluid undergoing a simple shear flow. A flow with a non-uniform velocity gradient, e.g. a Poiseuille flow, would introduce another length scale into the problem. Having three length scales in the problem, a microscopic length (colloidal particle size), the characteristic length of the external potential and a length characterizing the non-uniform flow, would make the present approach inapplicable.

Finally, we should note that, while we derived a formal microscopic expression for the effective temperature, Eq.
together with Eqs. (21, 26) and (27, 28), we did not calculate the effective temperature theoretically. To do this, one would have to develop a theoretical approach to calculate interaction contributions to the self-diffusion tensor (26) and the friction tensor (28). In an earlier work [9] one of us derived (but did not evaluate) mode-coupling expressions for these quantities. A different mode-coupling approach was proposed by Krüger and Fuchs [11]. These authors evaluated their expression for the effective temperature only in the vicinity of the mode-coupling transition. This fact makes a direct comparison of our simulational results with their predictions difficult.

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