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α-relaxation, shear viscosity, and elastic moduli of hard-particle fluids from a mode-coupling theory with a retarded vertex

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The recently proposed modification of the mode-coupling theory (MCT) in which the static structure used in the vertex is computed at a lower density than the actual one is tested on several dynamics-related properties. The predictions from this modified version of MCT calibrated on the one-component hard-sphere fluid are found in very good agreement with simulation data for one-component and binary hard-sphere fluids. They are also relevant for the stress moduli for models with attractive tails beyond the hard core. The clear improvement observed on several properties should give a new impetus to the use of MCT as a quantitative tool.

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I. INTRODUCTION

Mode-coupling theory (MCT) is one of the few first-principles approaches of classical time-dependent correlation functions, especially in condensed matter systems with slow dynamics [1–3]. As it incorporates the interactions between particles through the static structure, one may go beyond packing or topological frustration considerations that usually suffice [4–7] to describe jammed or arrested states due to steric forces. Specific aspects such as low-density arrested states in colloids [8–16] or metallic glasses (see, for example, Ref. [17]) can thus be studied within MCT. Decades of work have shown [1,2] that it provides at least a qualitative account of several aspects of the slowing down, such as the different relaxation regimes and the scaling behavior of the density time-correlation functions [18]. Its prediction of the nonergodicity parameter in the hard-sphere glass is even quantitatively accurate. The glass transition lines can also be determined by MCT, but it generally overestimates the extent of the nonergodicity domain. Together with some questions about the interpretation of the ideal glass transition [19,20] or the possibility to predict the dynamics from the static structure only [21], these inaccuracies cast doubt on the use of MCT as a quantitative tool, though it is still often used for a first estimation of several properties, beyond the arrest itself.

To improve its predictions, the source of its insufficiencies should be identified. The present consensus (see, for example. Refs. [22] and [23]) attributes them to exaggerated correlations, a consequence of the factorization ansatz, which replaces in the memory function four-point contributions by products of pair terms [1,3,24]. To correct this from first principles one should start from the exact time evolution equation from which MCT is derived. This requires one to close the equations at a higher order as first shown by Szamel [25]; see also Ref. [26] for an alternative approach. Though this is the route from which well-controlled improvements might arise, these fundamental developments are technically complex and remain to be fully tested (see Refs. [27] and [28] for a recent overview). The effort for developing simpler *ad-hoc* corrections that preserve the experience accumulated with the standard MCT equations thus remains valuable. That this is still a timely question is further justified by the difficulty of investigating slow dynamics by computer simulation. In this pragmatic point of view, Nauroth and Kob [22] proposed two simple corrections for the Lennard-Jones mixture, a cutoff in the q-integrals or using an effective temperature, while Flenner and Szamel [23] suggested that comparison between MCT and simulation should be best made at the same reduced temperature. In Ref. [29], Voigtmann *et al.* adjusted the packing fractions and the wave number at the peak of $S(q)$ to make MCT agree with simulation. Alternatively, a time-convolutionless mode-coupling approach has been formulated [30]. Concerning the transport properties, and following previous work by Götze *et al.* [1], Banchio *et al.* [31] found that rescaling the density dependence of the long-time self-diffusion coefficient was necessary to make the MCT shear viscosity of hard spheres agree with experiment. A similar scaling was performed in Ref. [32] for comparing the MCT arrest lines for the square-well fluid to simulation data.

In a similar spirit, we recently [33] implemented the idea of reducing correlations by using in the vertex a static structure computed at a lower density than the actual one. By calibrating the correction on the experimental critical density of colloidal hard spheres, we have shown in Ref. [33] that this idea works well in the long-time limit (see Ref. [34] for a preliminary report), as well as for some aspects of the time-correlation functions, even for models with attractive contributions to the interaction potential. By providing evidence that this modification is efficient well beyond the properties considered in Ref. [33], the purpose of this work is to highlight the possibility to resort to MCT for quantitative studies.

II. THEORY

MCT is well documented, and the reader may refer to the work of Nagele *et al.* [35] for a detailed presentation

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for colloidal systems. We briefly recall here the equations required for the modification. We start with the MCT time evolution of the matrix $\mathbf{S}(q;t)$ of coherent intermediate scattering functions with elements $S_{\alpha\beta}(q;t)$:

$$\frac{\partial}{\partial t}\mathbf{S}(q;t)+q^2\mathbf{H}(q)\mathbf{S}^{-1}(q)\mathbf{S}(q;t)+\int_0^t dt'\mathbf{M}(q;t-t')\mathbf{H}^{-1}(q)\frac{\partial}{\partial t'}\mathbf{S}(q;t')=0, \tag{1}$$

where the matrix elements of the irreducible collective memory function are [35]

$$M_{\mu\nu}(q,t)=\frac{D^0_\mu D^0_\nu}{16\pi^3(\rho_\mu\rho_\nu)^{(1/2)}}\sum_{\gamma\gamma'\delta\delta'}\int d\mathbf{k}V_{\mu;\gamma\delta}(\mathbf{q},\mathbf{k})V_{\nu;\gamma'\delta'}(\mathbf{q},\mathbf{k})S_{\gamma\gamma'}(\|\mathbf{q}-\mathbf{k}\|;t)S_{\delta\delta'}(k;t) \tag{2}$$

with the vertex amplitude

$$V_{\mu;\gamma\delta}(\mathbf{q},\mathbf{k})=\frac{1}{q}[\mathbf{q}\cdot\mathbf{k}\delta_{\mu\delta}C_{\mu\gamma}(k)+\mathbf{q}\cdot(\mathbf{q}-\mathbf{k})\delta_{\mu\gamma}C_{\mu\delta}(\|\mathbf{q}-\mathbf{k}\|)+q^2x_\mu^{1/2}C^{(3)}_{\mu\gamma\delta}(\mathbf{k},\mathbf{q}-\mathbf{k})]. \tag{3}$$

ρ_μ is the density of species μ and $C_{\mu\nu}=(\rho_\mu\rho_\nu)^{1/2}c_{\mu\nu}$, with $c_{\mu\nu}$ the Fourier transform of the two-body static direct correlation functions (and similarly with $C^{(3)}$). Neglecting hydrodynamic interactions, the matrix $\mathbf{H}$ of partial hydrodynamic functions is $H_{\alpha\beta}=D^0_\alpha\delta_{\alpha\beta}$ [35] with D^0_α the Stoke-Einstein diffusion coefficient of species α.

The modification is based on the view that the source of the excessive tendency to dynamical arrest is the progressive buildup of too strong correlations in the memory term. It thus consists in using effective (lower) densities ρ^{eff}_α in the vertex. The actual densities are retained in $M_{\mu\nu}$ in the factor before the integral, and to remain consistent with the short-time limit, the true static structure $\mathbf{S}(q)$ is used in the second term of Eq. (1). This version was labeled as $V2$ in Ref. [33]. As a result, the MCT equation is solved for the actual densities but with a static structure in the vertex computed for ρ^{eff}_α (similarly, an effective temperature T^{eff} can be introduced when temperature is a relevant variable). Since the vertex corresponds to a less correlated system, the dynamical slowing down should be deferred to higher density. It is, however, difficult to predict the precise impact of this modification, given the intricacy of $M_{\mu\nu}(q,t)$ and $V_{\mu;\gamma\delta}(\mathbf{q},\mathbf{k})$. In a related way, the idea of rescaling the MCT vertex by a multiplicative factor in the memory function was introduced by Fabbian *et al.* [36] in their study of linear molecules.

It is stressed that this differs from the procedure by which the results from the original MCT are represented *a posteriori* using scaled variables. Rather, it amounts to solving the MCT equation with a "density-retarded" vertex preserving thus its predictive status, independently of the considered specific property.

Technically, the solution of the modified Eq. (1) is the same as for the original theory, except for the necessity to compute the input static structure at the actual and the corrected density. For this, we mostly used the reference hypernetted chain (RHNC) closure with bridges functions obtained from Rosenfeld's hard-sphere functional [37] (see Ref. [33] for details). As usual, the calculation starts with the short-time limit, without the memory term in Eq. (1), in which case $\mathbf{S}(q,t)$ can be computed analytically from $\mathbf{S}(q)$. $\mathbf{S}(q,t)$ at all times is then computed following the algorithm of Fuchs *et al.* [38], as detailed in Ref. [23] in matrix form for mixtures. Standard discretization was used for the time intervals, which span several orders of magnitude, the diffusion time $\tau=\frac{D_1{}^2}{D_0}$ being used as the unit timescale (the hard-sphere (HS) diameter D_1 is taken as the unit length). For the q-integrals, $n_q=500$ mesh points and $dq=0.183/D_1$ was found sufficient.

The one-component hard-sphere fluid with packing fraction $\phi=\frac{\pi}{6}\rho\sigma^3$ is used to calibrate the correction, starting from the difference $\Delta\phi=\phi^{ex}_g-\phi^{\text{MCT}}_g$ between the "exact" critical glass packing fraction and the MCT one. We then solve the MCT equation at the packing fraction ϕ, using in the vertex $C=\rho_{\text{eff}}c(q;\phi^{\text{eff}})$ computed for an effective packing fraction

$$\phi^{\text{eff}}=\phi-\Delta\phi. \tag{4}$$

The experimental critical density for colloidal hard spheres [39], $\phi^{ex}_g=0.58$, is often taken as the reference for the hard-sphere glass (hereafter, "*ex*" will refer to computer simulation when no experimental data are available, and the results from the modified MCT are labeled with a tilde). For MCT, the value depends on the input static structure, giving, for instance, $\phi^{\text{MCT}}_g=0.516$, with $S(q)$ from the Percus Yevik (PY) closure and 0.525, with the Verlet-Weiss correction [40]. In Ref. [41] we also obtained the same value from the RHNC integral equation. Unless specified the results shown here have been computed with $\Delta\phi=0.055$, making the correction independent of simulation [the form of Eq. (4) appropriate for mixtures is given by Eq. (8) in Ref. [33]]. Note that Foffi *et al.* [42] used $S(q)$ from simulation and found $\phi^{\text{MCT}}_g=0.546$. Together with $\phi^{ex}_g=0.58$, this gives $\Delta\phi=0.034$ (see the discussion of Figs. 6 and 7 below).

III. RESULTS

Most of the material presented in Ref. [33] concerned the impact of the modification on the critical density. For the HS-binary mixture studied in Ref. [42], using $\Delta\phi=0.055$ we found $\tilde{\phi}^{\text{MCT}}_g=0.6113$ for system A: $\delta=0.6$, $x_1=0.2$, with $\delta=D_1/D_2$ the small-to-big sphere diameter ratio and $x_1=\phi_1/(\phi_1+\phi_2)$ the relative packing fraction of the small spheres. This value, in a definitive improvement over the original MCT, is barely distinguishable from the simulation one, $\phi^{ex}_g=0.605$, estimated in Ref. [42] from the α-relaxation data. The latter value is found with $\Delta\phi=0.050$ used for a similar plot in Ref. [33] (Fig. 5). Figure 1 shows the approach

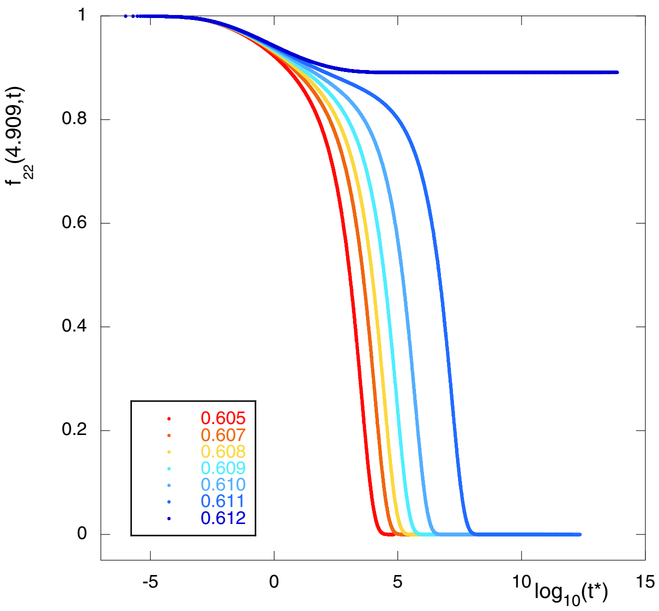

FIG. 1. Density autocorrelation function $f_{22}(q = \frac{4.909}{D_2}, \frac{t}{\tau_D})$ of the big spheres versus reduced time ($\log_{10}$ scale) in system A (computed with $\triangle\phi = 0.055$). The packing fractions indicated from top to bottom in the inset correspond to the figures from left to right.

of the dynamical arrest on the density correlators $f_{\alpha\beta}(q;t) = S_{\alpha\beta}(q;t)/S_{\alpha\beta}(q)$ of the big spheres and Figs. 2 and 3 the timescale determined by fitting their α-relaxation part by the Kohlrausch stretched exponential:

$$f_{ij}(q;t) = A_{ij} \exp\{-[t/\tau_{ij}(q)]^{\beta_{ij}(q)}\}. \tag{5}$$

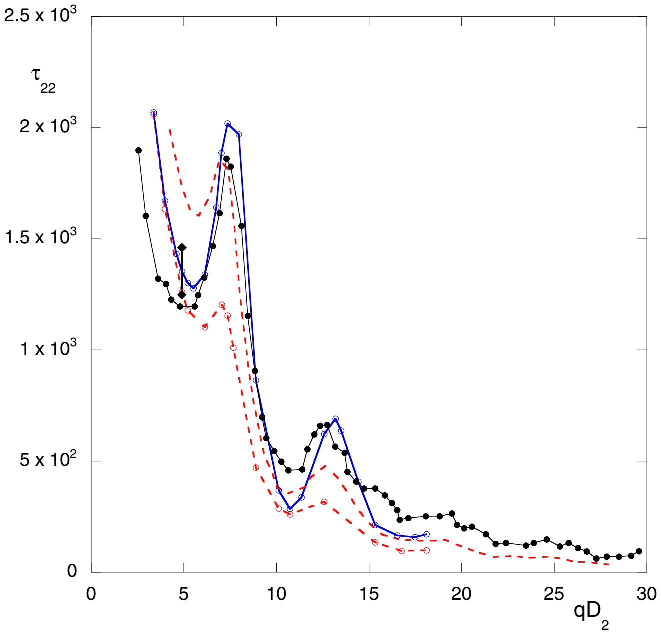

FIG. 2. α-relaxation timescale of the density autocorrelation function of the big spheres in system A. Solid lines: empty circles: corrected MCT, filled circles: simulation extracted from fig. 15 in Ref. [42]. Dashed lines: MCT with S_{ij} from simulation; empty circles: MCT with S_{ij} from the RHNC equations. The lines are guides for the eye.

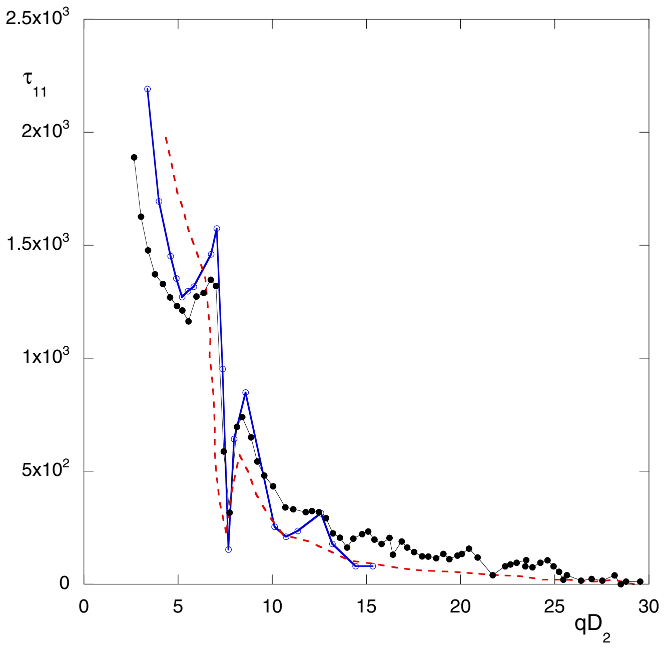

FIG. 3. Same as Fig. 2 for the small spheres (MCT and RHNC data not shown).

The corrected MCT is definitely in better agreement with simulation than the original MCT, when both use the RHNC static structure. On the whole, it is also better than when the "exact" static structure is used in the unmodified version. This is especially true for the timescale τ_{11} for the small spheres; see, for example, the maximum near $qD_2 = 7.5$, absent in the data from the unmodified version. The comparison was not pushed further due to the sensitivity of the fit to several factors such as the time interval [42]. The related uncertainty in the fit increases at large wave vector, due to the fact that the α-relaxation becomes less well defined, as shown in Fig. 4. The error bar shown for $qD_2 = 4.909$ used to convert the simulation time into MCT time for the same value of the correlator gives an idea of this sensitivity. Nevertheless, the results for τ_{22} and τ_{11} confirm that in addition to predicting a critical density that is nearly the simulation value, the modification does preserve the scaling properties of the MCT dynamical properties up to providing a nearly quantitative account of the relaxation timescales.

We now present the assessment of the proposed modification on a rheological property of the hard-sphere fluid, the zero-shear limiting viscosity η. We refer to the work of Banchio *et al.* [31] for the derivation of the necessary equations. The zero frequency viscosity is given by the relation

$$\eta = \eta'_\infty + \int_0^\infty dt \triangle\eta(t), \tag{6}$$

where η'_∞ is the high-frequency limiting viscosity, taken as in Ref. [31] from the Einstein form $\frac{\eta'_\infty}{\eta_0} = 1 + 2.5\phi$, with η_0 the solvent viscosity. $\triangle\eta(t)$ is the shear stress relaxation function

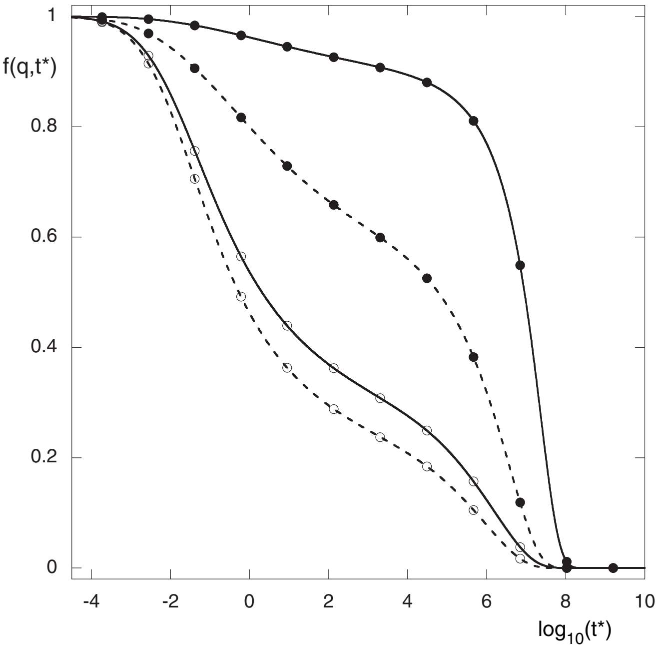

FIG. 4. Density autocorrelation functions $f_{ij}(\frac{q}{D_2}, \frac{t}{\tau_D})$ versus reduced time (log$_{10}$ scale) in system A. Filled circles: big spheres; empty circles: small spheres. Full curves: $qD_2 = 7.67$; dashed curves $qD_2 = 14.42$; $t* = \frac{t}{\tau_D}$.

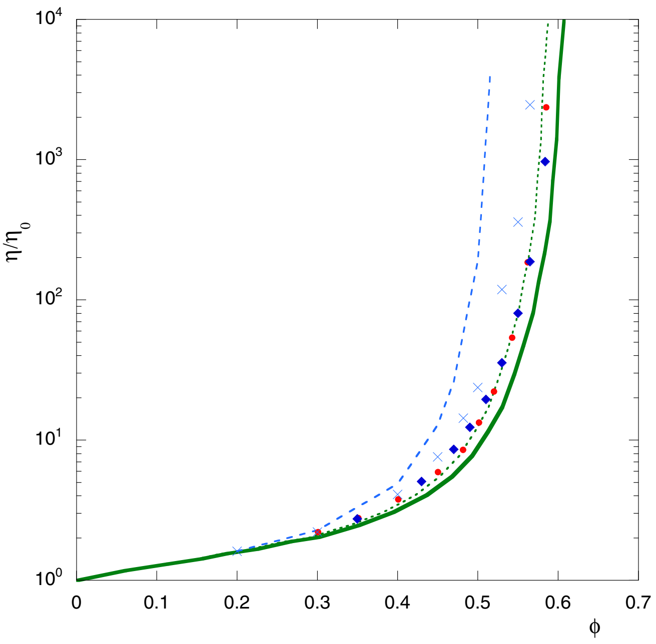

FIG. 5. Shear viscosity versus volume fraction of the hard-sphere fluid. Filled circles: BD simulation; diamonds: modified MCT with $\triangle\phi = 0.055$; crosses: same with $\triangle\phi = 0.043$. Dashes: original MCT, this work. Dotted line: viscosity versus rescaled volume fraction from Ref. [31] with $\phi_g = 0.6$. Thick solid line: same with $\phi_g = 0.62$.

given in the framework of MCT by [31] by

$$\triangle\eta(t) = \frac{k_BT}{60\pi^2}\int_0^\infty dqq^4\left[\frac{1}{S(q)}\frac{dS(q)}{dq}\right]^2\left[\frac{S(q;t)}{S(q)}\right]^2. \qquad (7)$$

The prediction for the viscosity η is a stringent test of the theory since both the time and the q dependence of the correlators are involved in Eqs. (6) and (7). Figure 5 shows the MCT predictions together with Brownian dynamics simulation data for colloidal hard spheres [43]. The agreement between both is excellent. A similar comparison is made in Fig. 3 in Ref. [31], by plotting the MCT data versus a rescaled density $\widetilde{\phi} = \phi\frac{\phi_g}{0.525}$. With $\phi_g = 0.62$ determined such that the MCT result for the long-time self-diffusion coefficient agrees with the Brownian dynamics (BD) results at high concentration, the agreement is fair. When using $\phi_g = 0.6$ it is as good as the one found with our modification. The latter value is somewhat closer to the experimental glass transition density $\phi_g = 0.58$ albeit not fully consistent with the observed divergence of the viscosity at $\phi_g = 0.62$. The latter scaling is, however, made *a posteriori*, as an *ad-hoc* representation of the actual MCT $\eta(\phi)$ curve, as this often occurs when comparing MCT and simulation data [32]. While these scalings share with our method the idea that for many properties the variation with the state variables predicted by MCT is correct, albeit at quantitatively incorrect values, they have a clearly distinct status as already stated.

As a final illustration of the effect of the modification we consider mechanical properties which now involve only the long-time limit of MCT. The first one is the longitudinal stress modulus m_0 whose expression in the hydrodynamic approximation obtained by taking the $t \to \infty$ and $q \to 0$ limits of the memory kernel (for a one-component system) is [44]

$$m_0 = \int_0^\infty dqf^2(q)\rho S(0)\left[\frac{qS(q)}{2\pi}\right]^2\left\{c^2(q) + \frac{2}{3}\left[q\frac{dc(q)}{dq}\right] + \frac{1}{5}\left[q\frac{dc(q)}{dq}\right]^2\right\}. \qquad (8)$$

The second one is the shear stress modulus G' whose expression is

$$G' = \frac{k_BT}{60\pi^2}\int_0^\infty dqq^4\left\{f(q)\frac{d\ln S(q)}{dq}\right\}^2. \qquad (9)$$

We first tested the correction on the short-range square-well fluid of relative width $\epsilon = 0.03$ investigated in Ref. [44]. Temperature is then relevant, but as shown in Ref. [33], a significant impact of the transition lines in the (T, ρ) plane is observed even when the correction is independent of temperature. Since this model does not correspond to an actual physical system and in the absence of simulation data, the effect of the correction was investigated only at the qualitative level with a temperature-independent correction, with the PY static structure and $\Delta\phi = 0.034$ for which the effect of the modification is less pronounced. The variations of the longitudinal stress modules m_0 and of the shear stress modulus G' along the the fluid-glass line are shown in Figs. 6 and 7. The corrected curve in Fig. 6 is qualitatively similar to the original one but shifted: the same rigidity is found at a higher density since, at fixed temperature, the glass occurs at a higher density than with the original theory. Equivalently,

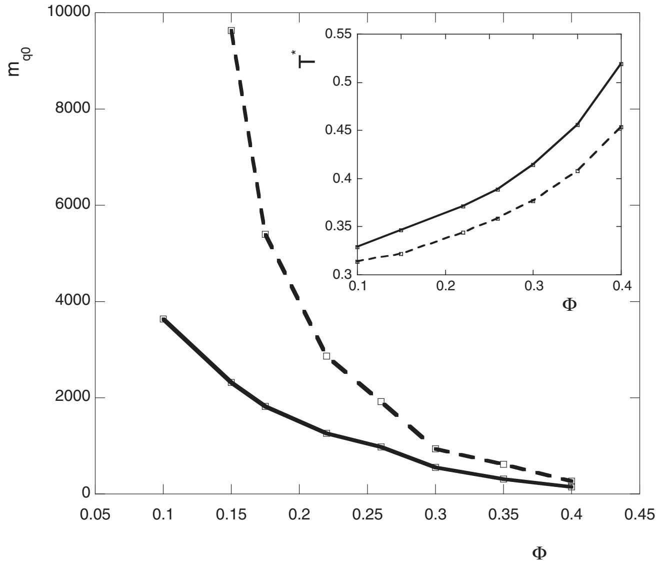

FIG. 6. Longitudinal stress modulus along the fluid-attractive glass line. Filled circles: original MCT with Percus-Yevick static structure as in Fig. 7 in Ref. [44]. Open circles: modified MCT. The lines are guides for the eye. The inset shows the corresponding transition lines.

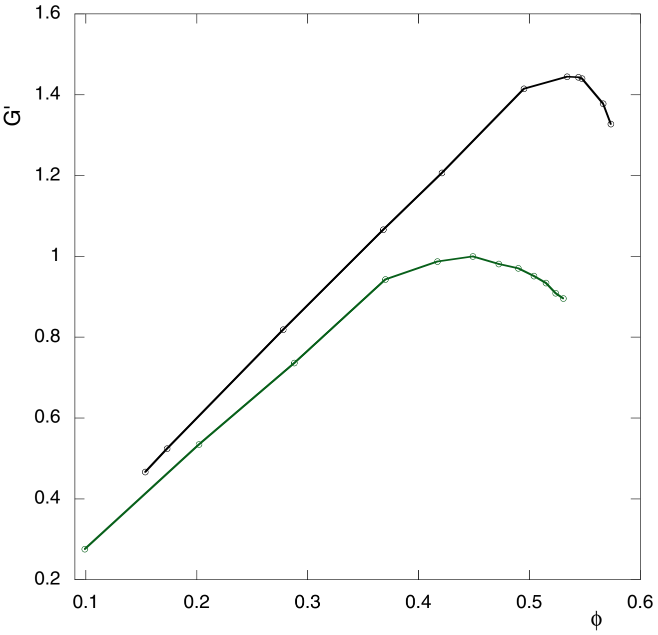

FIG. 7. Shear stress modulus along the fluid-attractive glass line (arbitrary units). Lower curve: original MCT as in Fig. 7 in Ref. [44]. Upper curve: modified MCT. Both use the Percus-Yevick static structure.

a given density corresponds to a lower transition temperature and hence a stiffer “bonded” glass. A similar effect is observed in Fig. 7. Our numerical values differ from those of Ref. [44], but the value of G' depends on the precise density at which it is computed on the glass side of the line. The data are thus shown in arbitrary units, to focus on the relative variation with respect to the original MCT. The first observation is the significant increase in relative magnitude of G' (recall the lower transition temperature with the modification). Next, the maximum stiffness with respect to shear [44] should be found at a higher density. We found a similar behavior with the Yukawa potential of width ϵ and inverse range b investigated in Ref. [8]. For example, for $k_BT/\epsilon = 0.12$ and $b = 20\sigma$ we find $G' \simeq 34$ and $G' \simeq 61$ (in the units of Fig. 5 in Ref. [8]) without and with correction, respectively. These observations are consistent with the main effect of the modification in the long-time limit: the critical density is higher. Studying in more detail the complex behavior with temperature, the difference between the shear and longitudinal stress moduli [44], including the repulsive glass, would, however, require a temperature-dependent correction. All these points need further clarification eventually by simulation, possibly on more specific models.

To conclude, this study confirms that the idea of tempering the buildup of correlations that are too strong in the MCT memory kernel by computing the vertex at a lower density leads to an overall improvement of the prediction of MCT both in the long-time limit and in the relaxation dynamics, up to providing a very good account of some rheological and mechanical properties. This clear gain in accuracy with very little cost and no *ad-hoc* scaling of the state variables is certainly useful from the applications point of view. On the other hand, while it brings no further insight on the fundamental mechanisms involved in the dynamical slowing down, it should be beneficial to the theory since it stresses again the importance of a proper treatment of the memory kernel. Its assessment on other models and properties should stimulate further theoretical and simulation work.

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