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Theory for the dynamics of glassy mixtures with particle size swaps

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We present a theory for the dynamics of binary mixtures with particle size swaps. The general structure of the theory shows that, in accordance with physical intuition, particle size swaps open up an additional channel for the relaxation of density fluctuations. Thus, allowing particle size swaps speeds up the dynamics. To make explicit predictions, we use a factorization approximation similar to that employed in the mode-coupling theory of glassy dynamics. We calculate an approximate dynamic glass transition phase diagram for an equimolar binary hard sphere mixture. We find that in the presence of particle size swaps, with increasing ratio of the hard sphere diameters the dynamic glass transition line moves towards higher volume fractions, up to the ratio of the diameters approximately equal to 1.2, and then saturates. We comment on the implications of our findings for the theoretical description of the glass transition.

Introduction. – Until recently, computer simulation studies of supercooled fluids suffered from the inability to equilibrate model glass-forming systems at temperatures close to those corresponding to the laboratory glass transition temperature [1]. Advances in the so-called swap dynamics computer simulation algorithms allowed researchers to overcome this restriction for a class of glass-forming systems [2, 3]. Swap dynamics algorithms add Monte Carlo moves in which exchanges of the diameters of two different particles are attempted to standard local Monte Carlo or Molecular Dynamics simulations. It has been found that the slow down of the swap dynamics algorithms with increasing density and/or decreasing temperature is much less drastic than that of the standard local Monte Carlo or Molecular Dynamics algorithms. Since swap dynamics generates the same equilibrium ensemble as either local Monte Carlo or Molecular Dynamics, more gradual slowing down makes possible equilibration of certain model systems at temperatures equal to or below those corresponding to the laboratory glass transition temperature, which enables studies of equilibrium properties of deeply supercooled fluids and glassy solids [4].

The advances in the swap dynamics algorithms lead to interesting theoretical questions. First, why is swap dynamics so much faster than "normal" dynamics? Second, does the swap dynamics speed-up have implications for the description of supercooled fluids dynamics and the glass transition? In particular, how can the difference between the dynamics without and with swaps be reconciled with the so-called Random First Order Transition (RFOT) framework, which connects slowing down upon approaching the glass transition with changes of static quantities, the configurational entropy and the static point-to-set correlation length [1, 5, 6]. Here we briefly comment on the former question; we will return to the latter one at the end of this Letter.

While intuitively it seems plausible that exchanges of particles' radii result in significant changes of local neighborhoods, which should speed up the relaxation of density fluctuations, a theoretical description of this speed-up is lacking. In a recent preprint Brito *et al.* [7] asserted, on the basis of general arguments applicable only to systems with continuous polydispersity, that allowing particle size swaps results in the decrease of the onset temperature for glassy behavior. However, their approach does not lead to a specific quantitative prediction for this change. Somewhat earlier, Ikeda *et al.* [8] used the correspondence between the so-called (avoided) dynamic glass transition observed in simulations [9] and predicted by approximate theories [10], and the dynamic transition predicted by replica theory [12]. These two transitions coincide in the only exactly solvable glassy particle-based model, the infinitely dimensional model of spherically symmetric particles [13, 14]. Ikeda *et al.* argued that the presence of the exchanges of particles' diameters implies a more general structure of the *Ansatz* for the inter-replica correlation for a simple mean-field-like glass-forming model, the binary Mari-Kurchan model [15, 16]. They showed that the new *Ansatz* allows one to distinguish between dynamic transitions without and with exchanges of particles' diameters.

In this Letter we present a *dynamic* theory for the acceleration due to the particle size swaps. First, on the basis of the general structure of the theory, we argue that particle size swaps open an additional relaxation channel, which speeds up the dynamics. Then, to make explicit predictions, we use a factorization approximation to evaluate irreducible memory functions [10]. We calculate an approximate dynamic glass transition phase diagram for an equimolar binary hard sphere mixture. In the presence of particle size swaps, with increasing ratio of the diameters the dynamic transition shifts towards higher volume fractions. The shift saturates at about 4% at the diameter ratio of approximately 1.2.

Model: binary mixture with particle size swaps. – We consider a binary mixture, which is the simplest model that allows one to investigate the influence of the particle size swaps on the dynamics. In a recent study of swap algorithms [3] it was found that particle size exchanges in systems with a continuous polydispersity result in the largest speed-up of the dynamics. Continuous polydispersity has some theoretical advantages [11], but there are no approximate expressions for equilibrium pair correlation functions for systems with continuous polydispersities, which makes explicit calculations difficult.

We consider a binary mixture consisting of N particles in volume V. Particles can be of type A or B, which differ by size. Since any particle can change its type, the state of particle i is determined by its position, $\mathbf{r}_i$, and type indicated by a binary variable σ_i, with $\sigma_i = 1$ corresponding to A and $\sigma_i = -1$ corresponding to B. The composition of the system is specified by the difference of the chemical potentials of particles of type A and B, $\Delta\mu$. We note that the composition depends also on the number density $n = N/V$ and the temperature T of the system. We will assume that these three parameters result in concentrations x_A and x_B, and that these concentration are constant while the density or the temperature of the system varies. In practical calculations we will restrict ourselves to equimolar mixtures (for which some formulas simplify). We assume that the "normal" dynamics of the system is Brownian, *i.e.*, that each particle moves under the combined influence of thermal noise and interparticle forces. The forces are derived from a spherically symmetric potential, which depends on the particle type, $V_{\sigma_i\sigma_j}(r_{ij})$, where $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ is the distance between particles i and j. In addition to the Brownian motion in space, the particles can change their type (size). We assume that each particle can change its type independently of the type changes of other particles. This is analogous to the single-spin-flip dynamics of spin systems. In contrast, in practical computational applications one typically changes the types/sizes of two particles in such a way that the number of particles of each size is conserved, which is analogous to the so-called Kawasaki dynamics of spin systems. The latter procedure is convenient because it allows one to maintain easily a specific composition of the system. We believe that replacing the latter procedure by single particle size swaps is a relatively mild change and that both procedures lead to qualitatively similar results.

The above described model corresponds to the following equation of motion for the N-particle distribution, $P_N(\mathbf{r}_1, \sigma_1, ..., \mathbf{r}_N, \sigma_N; t)$, abbreviated below as P_N,

$$\partial_t P_N = \Omega_{\mathrm{sw}} P_N \equiv (\Omega + \delta\Omega_{\mathrm{sw}})\, P_N. \tag{1}$$

Here the evolution operator Ω_{sw} consists of two parts describing two relaxation channels, the part describing Brownian motion of particles,

$$\Omega = D_0 \sum_i \partial_{\mathbf{r}_i} \cdot (\partial_{\mathbf{r}_i} - \beta \mathbf{F}_i) \tag{2}$$

and the part describing particle size swaps,

$$\delta\Omega_{\mathrm{sw}} = -\tau_{\mathrm{sw}}^{-1} \sum_i (1 - S_i)\, w_i. \tag{3}$$

In Eq. (2) D_0 is the diffusion coefficient of an isolated particle, $D_0 = k_B T/\xi_0$, with ξ_0 being the friction coefficient of an isolated particle, $\beta = 1/k_B T$, and $\mathbf{F}_i$ is the total force on particle i, $\mathbf{F}_i = \sum_{j\neq i} \mathbf{F}_{ij} = -\sum_{j\neq i} \partial_{\mathbf{r}_i} V_{\sigma_i\sigma_j}(r_{ij})$. In Eq. (3) τ_{sw}^{-1} is the rate of attempted particle size swaps, S_i is the swap operator, $S_i\sigma_i = -\sigma_i$, and w_i is the factor ensuring that the detailed balance condition, $(1 - S_i)\, w_i P_N^{eq} = 0$, is satisfied, with P_N^{eq} being the equilibrium distribution, $P_N^{eq} \propto \exp\left(-\beta \sum_{i\neq j} V_{\sigma_i\sigma_j}(r_{ij}) + \sum_i \frac{\beta}{2}\Delta\mu\sigma_i\right)$. The factor w_i depends on the way particle size swaps are attempted. In practical applications one typically uses Metropolis criterion for accepting attempted swaps. It should be emphasized that while the interactions influencing particles' motion in space are pairwise-additive, the factor w_i typically is not and it depends on the whole neighborhood of particle i.

The basic object of our theory are the density correlation functions,

$$F_{\alpha\beta}(q;t) = \left\langle n_\alpha(\mathbf{q}) e^{\Omega_{\mathrm{sw}} t} n_\beta(-\mathbf{q}) \right\rangle, \tag{4}$$

where n_α, $\alpha = A, B$, are the Fourier transforms of the normalized microscopic densities of particles of type α,

$$n_\alpha(\mathbf{q}) = \frac{1}{\sqrt{N}} \sum_i \frac{1 + \sigma_i\,(\delta_{\alpha A} - \delta_{\alpha B})}{2} e^{-i\mathbf{q}\cdot\mathbf{r}_i}. \tag{5}$$

In Eq. (4) and in the following equations the standard conventions apply: $\langle \ldots \rangle$ denotes the semi-grand canonical ensemble average over P_N^{eq}, the equilibrium probability distribution stands to the right of the quantity being averaged, and all operators act on it as well as on everything else.

We should emphasize that, in contrast to the approach of Ikeda *et al.*, in our theory the functions that characterize the dynamics and whose non-zero long-time limits signal the dynamic glass transition are the same for systems evolving without and with swap dynamics.

General theory. – We use the standard projector operator procedure to derive the general structure of the theory for the dynamics with particle size swaps. First, we define a projection operator on the density subspace, $\mathcal{P}$

$$\mathcal{P} = \cdots \sum_{\alpha\beta} n_\alpha(-\mathbf{q})\rangle\, S_{\alpha\beta}^{-1}(q)\, \langle n_\beta(\mathbf{q}) \ldots, \tag{6}$$

and the orthogonal projection, $\mathcal{Q}$,

$$\mathcal{Q} = \mathcal{I} - \mathcal{P} \equiv \mathcal{I} - \cdots \sum_{\alpha\beta} n_\alpha(-\mathbf{q})\rangle\, S_{\alpha\beta}^{-1}(q)\, \langle n_\beta(\mathbf{q}) \ldots. \tag{7}$$

In Eqs. (6-7) $S_{\alpha\beta}(q)$ denote the partial structure factors, $S_{\alpha\beta}(q) \equiv F_{\alpha\beta}(q; t = 0)$. Next, using projection operator identities [10, 17, 18] we express the Laplace transforms of the time-derivatives of the density correlation functions in terms of the *reducible* memory functions,

$$zF_{\alpha\beta}(q;z) - S_{\alpha\beta}(q) = -\sum_{\gamma\delta} \left(O_{\alpha\gamma}(q) - M_{\alpha\gamma}^{\mathrm{red}}(q;z)\right) S_{\gamma\delta}^{-1}(q) F_{\delta\beta}(q;z). \tag{8}$$

In Eq. (8) O is the frequency matrix, $O_{\alpha\beta}(q) = -\langle n_\alpha(\mathbf{q})\Omega_{\rm sw} n_\beta(-\mathbf{q})\rangle$ and $M^{\rm red}_{\alpha\gamma}(q;z)$ is the matrix of reducible memory functions. In the present case, with two different relaxation channels, it is convenient to express both matrices in terms of 3-dimensional vectors v_α, $\alpha = A, B$, and 3x3 matrices O and $\mathsf{M}^{\rm red}$,

$$O_{\alpha\beta}(q) = \mathsf{v}_\alpha^{\rm T}\mathsf{O}(q)\mathsf{v}_\beta, \tag{9}$$
$$M^{\rm red}_{\alpha\beta}(q;z) = \mathsf{v}_\alpha^{\rm T}\mathsf{M}^{\rm red}(q;z)\mathsf{v}_\beta. \tag{10}$$

Here $\mathsf{v}_A^{\rm T} = (1,0,1)$, $\mathsf{v}_B^{\rm T} = (0,1,-1)$, $\mathsf{O}_{11} = D_0 q^2 x_A$, $\mathsf{O}_{22} = D_0 q^2 x_B$, $\mathsf{O}_{33} = (1/2N\tau_{\rm sw})\left\langle\sum_i w_i\right\rangle$, $\mathsf{O}_{ab} = 0$ for $a \neq b$, and the matrix $\mathsf{M}^{\rm red}$ reads

$$\mathsf{M}^{\rm red}_{ab}(q;z) = \left\langle \pi_a(\mathbf{q})\left(z - \mathcal{Q}\Omega_{\rm sw}\mathcal{Q}\right)^{-1}\pi_b(-\mathbf{q})\right\rangle, \tag{11}$$

where

$$\pi_{1,2}(\mathbf{q}) = \frac{D_0}{\sqrt{N}}\mathcal{Q}\sum_i i\mathbf{q}\cdot(i\mathbf{q} - \beta\mathbf{F}_i)\frac{1\pm\sigma_i}{2}e^{-i\mathbf{q}\cdot\mathbf{r}_i}, \tag{12}$$
$$\pi_3(\mathbf{q}) = -\frac{1}{\sqrt{N}\tau_{\rm sw}}\mathcal{Q}\sum_i w_i\sigma_i e^{-i\mathbf{q}\cdot\mathbf{r}_i}. \tag{13}$$

As argued by Cichocki and Hess [17] and later, more generally, by Kawasaki [19], for systems with stochastic dynamics, memory functions analogous to $\mathsf{M}^{\rm red}$ can be reduced further by introducing the so-called irreducible evolution operator [20].

We define the irreducible evolution operator $\Omega^{\rm irr}_{\rm sw}$ as follows [18]

$$\begin{aligned}
&\Omega^{\rm irr}_{\rm sw} = \mathcal{Q}\Omega_{\rm sw}\mathcal{Q} \\
&-\sum_{\alpha\beta}\mathcal{Q}\Omega n_\alpha(-\mathbf{q})\rangle\langle n_\alpha(\mathbf{q})\Omega n_\beta(-\mathbf{q})\rangle^{-1}\langle n_\beta(\mathbf{q})\Omega\mathcal{Q} \\
&-\mathcal{Q}\delta\Omega_{\rm sw}\sigma(-\mathbf{q})\rangle\langle\sigma(\mathbf{q})\delta\Omega_{\rm sw}\sigma(-\mathbf{q})\rangle^{-1}\langle\sigma(\mathbf{q})\delta\Omega_{\rm sw}\mathcal{Q},
\end{aligned} \tag{14}$$

where $\sigma(\mathbf{q})$ is the microscopic composition field,

$$\sigma(\mathbf{q}) = n_A(\mathbf{q}) - n_B(\mathbf{q}) = \frac{1}{\sqrt{N}}\sum_i \sigma_i e^{-i\mathbf{q}\cdot\mathbf{r}_i}. \tag{15}$$

We note that reducible parts of $\mathcal{Q}\Omega_{\rm sw}\mathcal{Q}$ are removed separately for the two relaxation channels in our system.

Using definition (14) we can express $\mathsf{M}^{\rm red}$ in terms of matrix $\mathsf{M}^{\rm irr}$ whose elements are functions evolving with the irreducible evolution operator,

$$\mathsf{M}^{\rm red}(q;z) = \mathsf{M}^{\rm irr}(q;z) - \mathsf{M}^{\rm irr}(q;z)\mathsf{O}^{-1}(q)\mathsf{M}^{\rm red}(q;z), \tag{16}$$

where

$$\mathsf{M}^{\rm irr}_{ab}(q;z) = \left\langle \pi_a(\mathbf{q})\left(z - \Omega^{\rm irr}_{\rm sw}\right)^{-1}\pi_b(-\mathbf{q})\right\rangle. \tag{17}$$

The combination of Eqs. (8-10) and (16-17) defines the general structure of our theory. To appreciate its meaning it is instructive to consider an approximation that neglects the time-delayed coupling between "normal" dynamics and particle size swaps. To this end we set $\mathsf{M}^{\rm irr}_{ab}(q;z) = 0$ for $a = 1,2$ and $b = 3$, and $a = 3$ and $b = 1,2$. One can show [18] that in this case the right-hand-side of Eq. (8) becomes a sum of two independent terms (implying two *parallel* relaxation channels), the first one originating from "normal" dynamics and the second one due to particle size swaps. If the relaxation rate due to the first term becomes very small, the presence of the second channel can dramatically speed up the dynamics. We should remember, however, that particle size swaps alone cannot equilibrate the system.

Mode-coupling-like approximation. – To make explicit predictions we need to calculate the elements of matrix $\mathsf{M}^{\rm irr}$. To this end we follow the spirit of the mode-coupling theory [10], which is one of the most-successful but also most-criticized theories for glassy dynamics in three dimensions. Specifically, we use the sequence of three approximations [10, 21]: we project functions π_a onto the subspace spanned by the parts of density products orthogonal to the one-particle densities, factorize four-point dynamic correlation functions while replacing the irreducible evolution operator by the original unprojected evolution operator, factorize four-point static correlation functions, and use some additional approximations that amount to neglecting higher-order correlation functions in the expressions for the so-called vertices [18]. In this way we obtain the following approximate expressions for the matrix elements of $\mathsf{M}^{\rm irr}$:

$$\begin{aligned}
&\mathsf{M}^{\rm irr}_{ab}(\mathbf{q};t) \approx \frac{1}{2}\sum_{\alpha,\ldots,\theta}\sum_{\mathbf{k}_1,\mathbf{k}_2}\langle\pi_a(\mathbf{q})n_\alpha(-\mathbf{k}_1)n_\beta(-\mathbf{k}_2)\rangle \\
&\times S^{-1}_{\alpha\gamma}(\mathbf{k}_1)S^{-1}_{\beta\delta}(\mathbf{k}_2)F_{\gamma\epsilon}(\mathbf{k}_1;t)F_{\delta\zeta}(\mathbf{k}_2;t) \\
&\times S^{-1}_{\epsilon\eta}(\mathbf{k}_1)S^{-1}_{\zeta\theta}(\mathbf{k}_2)\langle n_\eta(\mathbf{k}_1)n_\theta(\mathbf{k}_2)\pi_b(-\mathbf{q})\rangle.
\end{aligned} \tag{18}$$

The vertices originating from Brownian dynamics part of the evolution operator have the same form as in the standard mode-coupling theory for binary mixtures [10, 18]. For *an equimolar binary mixture* the new vertex, which originates from the particle size swaps, reads

$$\begin{aligned}
&\langle n_\eta(\mathbf{k}_1)n_\theta(\mathbf{k}_2)\pi_3(-\mathbf{q})\rangle = -\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{q}}\frac{\langle\sum_l w_l\rangle}{2N^{3/2}\tau_{\rm sw}}\sum_\mu \\
&\left[\sum_\lambda\left(S_{\eta\lambda}(k_1)\delta_{\theta\mu} + S_{\theta\lambda}(k_2)\delta_{\eta\mu}\right) - 4S_{\eta\mu}(k_1)S_{\theta\mu}(k_2)\right] \\
&\times(\delta_{\mu A} - \delta_{\mu B}).
\end{aligned} \tag{19}$$

For non-equimolar mixtures there are additional terms in the new vertex, which will be reported elsewhere. We note that to derive expression (19) we factored out the average $\langle\sum_i w_i\rangle$ [18]. This somewhat technical step results in the dynamic glass transition being independent of the detailed form of w_i.

The exact memory function representation of the density correlation function, Eq. (8) together with Eqs. (9-10, 16), combined with the approximate form of $\mathsf{M}^{\rm irr}$, Eqs. (18-19), constitute our mode-coupling-like theory for the dynamics of a binary mixture with particle size

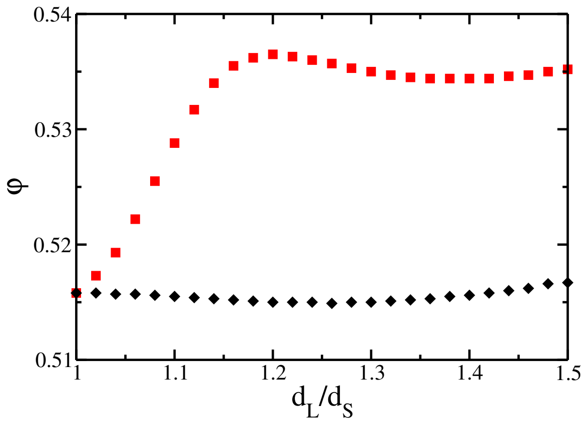

FIG. 1: Volume fraction φ at the dynamic glass transition as a function of the ratio of the hard sphere diameters d_L/d_S, for an equimolar binary hard-sphere mixture. Red squares and black diamonds denote the location of the transition with and without particle size swaps.

swaps. To get the explicit predictions for the time-dependence one has to solve these equations numerically.

Dynamic glass transition. – To find the location of the dynamic glass transition we assume that as the density increases and/or the temperature decreases density correlation functions develop plateaus and at the transition these plateaus do not decay. Thus, at the transition non-zero long-time limits $F_{\alpha\beta}(q;\infty)$ appear discontinuously. This assumption allows us to derive from our theory the following self-consistent equation for $F_{\alpha\beta}(q;\infty)$,

$$F_{\alpha\beta}(q;\infty) - S_{\alpha\beta}(q) = \\ -\sum_{\gamma\delta} \mathsf{v}_\alpha^{\mathrm{T}} \mathsf{O}(q) \left[\mathsf{M}^{\mathrm{irr}}(q,\infty)\right]^{-1} \mathsf{O}(q) \mathsf{v}_\gamma S^{-1}_{\gamma\delta}(q) F_{\delta\beta}(q;\infty). \tag{20}$$

where $\mathsf{M}^{\mathrm{irr}}(q,\infty)$ is given by Eq. (18) with non-zero long-time limits $F_{\alpha\beta}(q;\infty)$ substituted at the right-hand-side.

We solved self-consistent equations (20) for an equimolar binary hard-sphere mixture using as the static input approximate equilibrium correlation functions obtained from the Percus-Yevick closure [22–24]. As shown in Fig. 1, we found that for a system with particle size swaps the volume fraction at the dynamic glass transition increases with increasing particle diameter ratio up to the ratio of about 1.2, where the relative increase is about 4%, and then saturates. In agreement with Ref. [25], the location the dynamic glass transition for an equimolar mixture without swaps depends very weakly on the ratio of particle diameters for the ratio smaller than 1.5. In contrast, Ikeda *et al.* [8] found that the volume fraction at the dynamic transition both without and with particle size swaps increases with the ratio of particle diameters; the absolute separation of these transitions increases monotonically but the relative separation saturates, although at a diameter ratio larger than found here [26]. The relative difference at the diameter ratio of 1.2 is approximately the same according to both approaches. We note that the MK model considered by Ikeda *et al.* lacks non-trivial local structure but it is not clear whether this is the origin of the difference between our results and theirs. Finally, we show in the Supplemental Material that neglecting the coupling between "normal" dynamics and particle size swaps results in a phase diagram qualitatively similar to that showed in Fig. 1.

Discussion. – Wyart and Cates [27, 28] have argued that the success of swap dynamics algorithms in equilibrating model systems at temperatures comparable to the laboratory glass transition temperature implies that the RFOT scenario needs to be re-evaluated. They conjectured that the dominant barriers for low temperature relaxation are local and the growing static correlation length is responsible for only a small fraction of slowing down. In contrast, Ikeda *et al.* [8] argued that while the RFOT framework is still valid, there is a need for a more general way to calculate the configurational entropy. Effectively, they advocated using a less restrictive constrained equilibrium construction in the derivation of the replica theory for systems with particle size swaps. This is equivalent to their more general *Ansatz* for inter-replica correlations.

In our approach, the basic functions that signal the dynamic glass transition are the same for both "normal" and swap dynamics. Thus, we cannot account for the presence of particle size swaps by using a different *Ansatz* for our non-ergodicity parameters, $F_{\alpha\beta}(q;\infty)$. On the other hand, allowing for particle size swaps does change the location of the dynamic transition and influences the values of non-ergodicity parameters. We suggest that a possible way out of this conundrum is to recognize the fact that metastable states, whose appearance triggers the dynamic glass transition and which are counted by the configurational entropy, should be defined using a dynamical criterion. Some time ago [29] we showed that the standard mode-coupling theory's equation for the non-ergodicity parameter can be re-derived from a replica approach combined with a dynamic criterion (vanishing of a current). It would be interesting to check whether Eq. (20) of the present theory can be re-derived in a similar way [30]. We note that it is possible that different dynamics lead to equivalent definitions of metastable states (*e.g.* within mode-coupling theory Newtonian and Brownian dynamics result in the same dynamic glass transition scenario [21]). However, a significant modification of the dynamics may result in different states being metastable and different dynamic glass transition and configurational entropy scenarios. We leave these important issues for future work.

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