Revisiting the replica theory of the liquid to ideal glass transition

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The replica theory of the "Random First Order Transition" (RFOT) from a supercooled liquid to an "ideal" glass of a system of "soft spheres" is revisited. Following the seminal work of Mézard and Parisi (J. Chem. Phys. **111**, 1076 (1999)), the number m of weakly interacting replicas of the system is varied continuously from $m = 2$ to $m < 1$. Relevant order parameters and the free energy of the liquid and glass phases are calculated using the hyper-netted (HNC) approximation for the pair correlation functions. The scenario observed for all m confirms the existence of two glass branches G_1 and G_2. The latter has the lowest free energy for all $m > 1$, while the former has a lower free energy for $m < 1$, but is shown to be unstable against spinodal decomposition for any non-zero value of the attractive inter-replica coupling. The critical temperature T_{cr} of the RFOT turns out to depend on m, which may reflect the thermodynamic inconsistency of the HNC closure. The RFOT is predicted to be weakly first order, characterized by a small jump in density between the coexisting liquid and G_2 phases for all $m > 1$. Estimating T_{cr} in the limit $m \longrightarrow 1$ requires a proper extrapolation of high resolution HNC calculations. The present protocol allows a direct access to the free energy of the ideal glass phase below T_{cr}.

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I. INTRODUCTION

Our understanding of the transition of a supercooled liquid to an amorphous solid, or glass, has evolved dramatically over the last two decades. A generic scenario for fragile structural glasses (whose viscosity does not follow a simple Arrhenius behaviour as a function of temperature [1]) is by now generally accepted (for extensive reviews, see e.g. [2] and [3]). According to that scenario, when a liquid is rapidly cooled (i.e. quenched) below its equilibrium freezing temperature $T_{\rm f}$, it undergoes a succession of dynamical and thermodynamic transformations towards an "ideal" glass phase, a state which is thermodynamically stable after an infinitely long relaxation time, and hence experimentally unobservable. The successive stages of the scenario upon lowering the temperature are the following:

- A "kinetic" glass transition (GT) at a temperature $T_{\rm k} < T_{\rm f}$, characterized by a dramatic structural slowing down, as detected by inelastic neutron scattering experiments [4], and predicted by "mode coupling" theory (MCT) [5].

- A "laboratory" GT, at a temperature $T_{\rm g}$, where relaxational and experimental time scales become comparable; the latter are typically of the order of 10^3 sec. $T_{\rm g}$ is not an intrinsic property of the glass former, but depends on the quench rate: the slower the cooling rate, the lower $T_{\rm g}$. The "laboratory" GT is signalled by a rapid change in slope of the measured thermodynamic properties, like the molar volume [6].

- Below $T_{\rm g}$, "aging" of the material sets in, i.e. measured time-dependent correlation functions are no longer time-translational invariant, signalling that the system has not reached thermodynamic equilibrium. Moreover the local molecular dynamics are increasingly heterogeneous ("dynamical heterogeneity"), with quiescent (or frozen) nano-scale regions coexisting with regions undergoing structural rearrangement at any instant of time [7].

- It has long been conjectured that the deeply supercooled liquid finally undergoes a "random first order transition" (RFOT) to the "ideal" glass phase at some critical temperature $T_{\rm cr}$. On the basis of formal analogies with the mean-field predictions of some classes of spin glasses (corresponding to infinite dimensionality), it is generally accepted that $T_{\rm cr} = T_{\rm K}$ for structural glasses as well [8, 9], where $T_{\rm K}$ is the Kauzmann temperature [10] at wich the configurational entropy, related to the number of accessible metastable thermodynamic states of the system, vanishes. It must be remembered that the "ideal" glass is a "virtual" phase, since it requires an infinitely long relaxation process.

In an "ideal glass", atoms vibrate around disordered equilibrium positions ($\{\mathbf{R}_i\}$; $1 < i < N$), rather than around the periodic positions of a crystal lattice. In this disordered state, the system is "stuck" in the lowest minimum of the suitability defined free energy "landscape" [11]. Although a detailed characterization of such a landscape with an exponentially large number of local minima is clearly unachievable, the replica method for the study of disordered systems [12] can be put to good use to probe the landscape by defining a suitable order parameter [13, 14] which can be calculated, at least approximately, using techniques borrowed from the theory of liquids [15]. In a series of papers we recently used the replica approach to investigate a system of two weakly coupled replicas to locate the RFOT of systems of soft spheres [16] and of Lennard-Jones atoms [17]. Based on the hyper-netted chain (HNC) free energy functional [18] the calculations predicted that the RFOT is characterized by a discontinuity of the order parameter in the limit of vanishing inter-replica coupling, as expected, and by a small (1 percent) relative volume change between the liquid (L) and ideal glass G_2 phases at the critical temperature $T_{\rm cr}$. An independent estimate of the Kauzmann temperature $T_{\rm K}$ suggested that $T_{\rm cr} > T_{\rm K}$, confirming that the RFOT preempts the "Kauzmann catastrophe", i.e. a vanishing configurational entropy. The scenario predicted by HNC theory was confirmed by calculations on more advanced thermodynamically self-consistent integral equations [16, 17], although with significant quantitative differences. In a seminal paper, Mézard and Parisi [19] used the same HNC free energy functional which they extended to non-integer, continuously varying the number m of coupled replicas (as opposed to the more "pedestrian" choice $m = 2$ [14, 16, 17]). They determined $T_{\rm cr}$ (assumed to coincide with $T_{\rm K}$) in the range $m > 1$. In order to calculate the thermodynamic properties of the "ideal" glass below $T_{\rm cr}$, they extended their search to $m < 1$ and showed that these properties could be related to those of the supercooled liquid at the effective temperature T/m.

Their work was extended recently to explore the influence of critical correlations on slow relaxation in glasses, in the vicinity of the RFOT [20]. In the present paper, we adopt the strategy of continuously varying the number m of weakly coupled replicas within the framework of HNC theory [19], to look for liquid-like and glass-like solutions in the range $1 < m < 2$, using the search protocols previously applied to the $m = 2$ case [16, 17]. The present investigation is restricted to the well documented model of "soft spheres" [21]. The two key questions which will be addressed are:

a) Is the RFOT temperature $T_{\rm cr}$ independent of m, as implicitly assumed in Ref. [19] ?

b) Is the RFOT a weakly first order thermodynamic transition, as for $m = 2$ [16] or is it a second order transition, as assumed in Ref. [19], in the limit $m \longrightarrow 1$?

The outline of the paper is the following: Section 2 briefly recalls the model, the order parameters, the HNC approximation and the replica strategy. In section 3, the range $2 \geq m > 1$ is systematically investigated. The limit $m \longrightarrow 1$ is examined in section 4. The range $m < 1$ is explored in section 5. Conclusions are drawn in section 6.

II. THEORETICAL BACKGROUND

The "soft sphere" model is a three-dimensional system of atoms interacting via the pair potential

$$v(r) = \epsilon \left(\frac{\sigma}{r}\right)^n , \tag{1}$$

with the exponent n chosen here to be 12; ϵ sets the energy scale while σ is the atomic diameter. The advantage of this family of potentials lies in the fact that the excess (non-ideal) thermodynamic properties depend only on a single dimensionless parameter Γ, rather than on density ρ and temperature T separately [21] ; for $n = 12$:

$$\Gamma = \frac{\rho\sigma^3}{(k_B T/\epsilon)^{1/4}} = \frac{\rho^*}{(T^*)^{1/4}}, \tag{2}$$

such that:

$$\beta v(r) = \frac{\Gamma}{r^{12}}, \tag{3}$$

where $\beta = 1/k_B T$ and distances are measured in units of σ. Consider now a system of m identical replicas (or "clones"), the atoms of which are weakly coupled by a short-ranged inter-replica attraction chosen to be of the form:

$$\beta v'(r) = -\epsilon' \left[\frac{c^2}{r^2 + c^2}\right]^6 = -\epsilon' w(r), \tag{4}$$

The range parameter c is chosen to be 0.3 (in units of σ) to ensure that an atom of one replica can interact at most with one atom of another replica. The exact form of the chosen $v'(r)$ is irrelevant, since we shall be interested in the limit $\epsilon' \longrightarrow 0$. Such a symmetric m component "mixture" is characterized by only two pair correlation functions: the intra-replica correlation function $h(r)$ and the inter-replica correlation function $h'(r)$, whatever the value of m. The pair correlation functions are related to the corresponding direct correlation functions $c(r)$ and $c'(r)$ via the Ornstein-Zernike (OZ) relations [15]:

$$h(r) = c(r) + \rho c(r) \otimes h(r) + (m-1)\rho c'(r) \otimes h'(r), \tag{5a}$$
$$h'(r) = c'(r) + c(r) \otimes h'(r) + \rho c'(r) \otimes h(r) + (m-2)\rho c'(r) \otimes h'(r), \tag{5b}$$

where $\otimes$ denotes a three-dimensional convolution product. These relations must be supplemented by approximate closure relations; the HNC closure relations read [15]:

$$g(r) = \exp\left[-\beta v(r) + \gamma(r)\right], \tag{6a}$$
$$g'(r) = \exp\left[-\beta v'(r) + \gamma'(r)\right]. \tag{6b}$$

The great advantage of the HNC approximation over more advanced closures is that the excess Helmholtz free energy per particle βf_{ex} can be expressed in terms of the pair correlation functions alone [18, 22]:

$$\begin{aligned}\beta f_{ex} =& \frac{1}{2\rho m(2\pi)^3}\int d\mathbf{k}\{m\rho\hat{h}(k) \\ &- (m-1)\log(1+\rho\hat{h}(k) - \rho\hat{h}'(k)) - \log(1+\rho\hat{h}(k) + (m-1)\rho\hat{h}'(k))\} \\ &+ \frac{\rho}{2}\int d\mathbf{r}\{g(r)\left[\log g(r) + \beta v(r)\right] - h(r) - \frac{h^2(r)}{2}\} \\ &+ \frac{(m-1)\rho}{2}\int d\mathbf{r}\{g'(r)\left[\log g'(r) + \beta v'(r)\right] - h'(r) - \frac{h'^2(r)}{2}\}\end{aligned} \tag{7}$$

where $\hat{p}(k)$ denotes the dimensionless Fourier transform (FT) of any function $p(r)$:

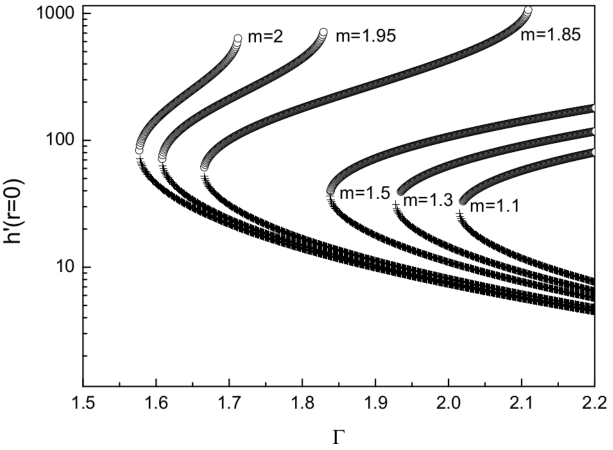

FIG. 1. Order parameter $h'(r=0)$ versus Γ for m =2.0, 1.95, 1.85, 1.50, 1.30 and 1.10.

$$\hat{p}(k) = \rho \int e^{-i\mathbf{k}\cdot\mathbf{r}} p(r) d\mathbf{r}, \tag{8}$$

All other thermodynamic properties then follow by taking appropriate derivatives of (7). In the rest of the paper, f^*_{ex} $=f_{ex}/\epsilon$ denotes the reduced excess free energy per particle.

The thermodynamic properties of the supercooled liquid L phase are calculated by setting $\epsilon' = 0$, i.e. by considering fully decoupled clones, implying $h'(r)$ $=c'(r) = 0$. For finite values of ϵ', the attraction between atoms of different clones will favour configurations corresponding to the same local free energy minimum. This tendency can be quantified by calculating the order parameter Q defined as the statistical average of the overlap function $q_{1,2}$ [14, 16] between two clones:

$$q_{1,2}(\{\mathbf{r}^1_i\}, \{\mathbf{r}^2_j\}) = \frac{1}{N}\sum_{i=1}^{N} w\left(\left|\mathbf{r}^1_i - \mathbf{r}^2_i\right|\right), \tag{9}$$

where $\mathbf{r}^1_i$ and $\mathbf{r}^2_i$ are the position vectors of the N particles of two different clones, 1 and 2.

$$Q = \langle q_{1,2}\rangle = 4\pi\rho \int_0^\infty g'(r) w(r) r^2 dr. \tag{10}$$

An alternative order parameter is provided by $h'(r=0)$. In the absence of inter-replica coupling $\epsilon' = 0$, $h'(r = 0) = 0$, while for finite ϵ', a central peak of the inter-replica correlation function $h'(r)$ is expected to build up as T is lowered (i.e. Γ increases) [16]. The two order parameters provide the key "diagnostics" to locate the RFOT. The search strategy, for a given value of Γ, is to solve the set of HNC equations for $h(r)$ and $h'(r)$, starting from an initial finite value of the inter-replica coupling ϵ', and to monitor the values of the order parameters Q and $h'(r = 0)$ upon gradually reducing ϵ'. If in the limit $\epsilon' \longrightarrow 0$, the order parameters take on their "random" (or un-correlated) values Q_r (as calculated from Eq. (9b) with $g'(r) = 1$) and $h'(r = 0) = 0$, the thermodynamic state Γ corresponds to the L phase. If, on the other hand, the order parameters take on values $Q >> Q_r$ and $h'(r = 0) >> 0$ in the limit $\epsilon' \longrightarrow 0$, the system is in a glass phase, where the replicas are all trapped within the same minimum of the free energy landscape. The process can be repeated for many values of the thermodynamic state parameter Γ, and a free energy versus Γ curve can be mapped out using Eq. (7), both for the supercooled liquid and glass phases. In fact our earlier calculations for $m = 2$ [16, 17] point to the existence of two glass phases, the stable, "ideal" glass phase G_2, and a metastable glass phase G_1 (of higher free energy). In the following sections we report our results for a continuously varying number $m \leq 2$ of clones.

In practice the coupled HNC equations for $h(r)$ and $h'(r)$ were solved numerically, using Gillan's very efficient iterative algorithm [23], for several resolutions Δr of the spatial grid and a corresponding variation of the total number N_g of grid points such as to keep the overall range in direct and reciprocal space large enough, and the grid spacing small enough, to minimize truncation and discretization errors of the numerical integrals. In practice, in the calculations N_g was never smaller than 4097 and Δr not larger than 0.01.

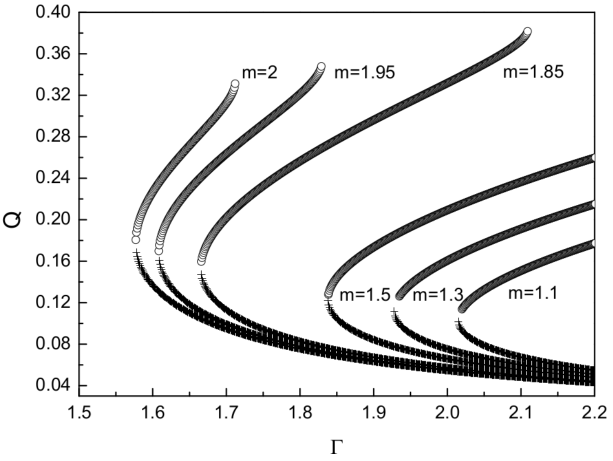

FIG. 2. Same as Fig. 1, but for the overlap parameter Q.

III. REDUCING THE NUMBER OF REPLICAS FROM $m = 2$ TO $m = 1.1$

This section presents results, using accurate HNC calculations, of the pair structure and RFOT order parameters $h'(r = 0)$ and Q, to map out the free energies of the L, G_1 and G_2 phases as functions of the soft sphere coupling parameter Γ, upon gradually reducing the number of replicas from $m = 2$ to $m = 1.1$. In practice, for given values of m and of the intra-replica coupling Γ, an initial value of the inter-replica coupling $\epsilon' = 0.001\epsilon$ is chosen; the variation of the order parameters $h'(r = 0)$ and Q is then monitored upon gradually reducing ϵ'.

Fig 1 shows the variation of $h'(r = 0)$ with Γ for six values of m. As Γ is lowered the order parameter increases along the G_1 branch, down to a threshold value, where a discontinuous jump occurs towards the G_2 branch along which the order parameter increases rapidly as Γ is increased. The latter behaviour is that expected for an "ideal glass" trapped in the lowest free energy minimum. The confinement of different replicas within this state is gradually enhanced as the temperature is lowered as reflected in the rapid increase of the order parameter. To the contrary, the increase of the order parameter as Γ is lowered, as observed along the G_1 branch is incompatible with the "ideal glass" behaviour. It is tentatively interpreted as a "defective" metastable state, where different replicas are trapped in neighbouring minima of the free energy hypersurface, separated by a free energy barrier; the latter is more easily overcome as the temperature increases [16, 17]. The metastable nature of the G_1 branch is confirmed by its systematically higher free energy compared to that the G_2 phase (cf Fig. 3).

Fig. 2 shows the corresponding results for the overlap order parameter Q. The overall scenario is identical to that revealed by Fig. 1, with quantitative agreement as regards the location of the discontinuous jumps from the G_1 to the G_2 phase. The differences in reduced excess free energies (per particle), $\Delta f^*_{ex}(\Gamma; m) = f^{*L}_{ex}(\Gamma) - f^{*G_1}_{ex}(\Gamma; m)$ and $f^{*L}_{ex}(\Gamma) - f^{*G_2}_{ex}(\Gamma; m)$ are plotted as functions of Γ for four values of m in Fig. 3. The free energy curves of the L and G_2 phases are seen to intersect at increasing critical values Γ_{cr} of Γ as the number m of replicas is continuously lowered. The intersection at Γ_{cr} signals the RFOT which appears to be a weakly first order thermodynamic transition characterized by a small discontinuity of the molar volume, in agreement with our earlier predictions for $m = 2$ [16, 17]. However the difference in slope of the L and G_2 free energy curves gradually decreases upon reducing m, pointing to the possibility of the weakly first order transition eventually turning into a second order transition as $m \longrightarrow 1$. This will be the subject of the following section. Table I summarizes the key predictions of our HNC calculations for $2 \geq m \geq 1.1$, namely the values of Γ at which the discontinuous jump of the order parameters $h'(r = 0)$ and Q between the G_1 and G_2 phases is observed, and the location of the intersection between the free energies of the G_2 and L phases, which we identify as the RFOT.

IV. APPROACHING THE LIMIT $m = 1$

This section focuses on the physically relevant limit $m \longrightarrow 1$. In this regime the differences in free energy between the three phases tend to become very small over an increasingly large range of couplings Γ, thus requiring an increasingly high spatial resolution of the HNC calculations.

Fig. 4 shows the dependence on Γ of the difference in reduced excess free energy of the L and G_2 phases:

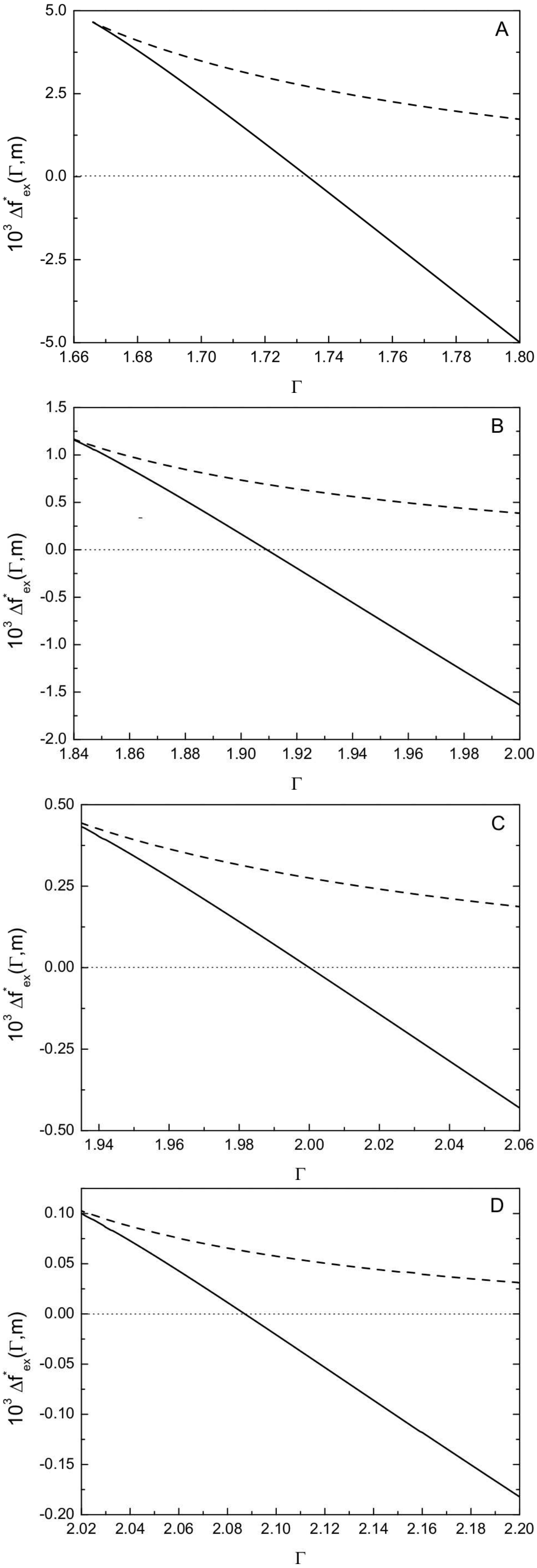

FIG. 3. Difference (multiplied by 10^3) in reduced excess free energies (per particle) of the L and G_1 phases (dashed line) and of L and G_2 phases (full line) for m =1.85 (A), 1.50 (B), 1.30 (C) and 1.10 (D).

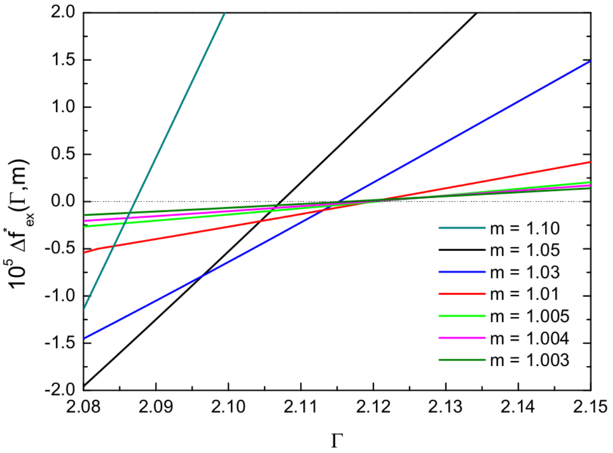

FIG. 4. Difference in reduced excess free energies (per particle) of the L and G_2 phases (multiplied by 10^5) for several values of m (listed in the inset).

$$\Delta f^*_{ex}(\Gamma; m) = f^{*L}_{ex}(\Gamma) - f^{*G_2}_{ex}(\Gamma; m), \tag{11}$$

for $1.1 \geq m \geq 1.003$. As m decreases, the RFOT, which corresponds to $\Delta f^*_{ex} = 0$, is shifted to higher critical values Γ_{cr}. Eventually, for m <1.005, Γ_{cr} ceases to increase and "sticks" to the value Γ_{cr}=2.12, which may hence be identified as the location of the RFOT in the limit $m \longrightarrow 1$. In view of the extremely small values of Δf^*_{ex} over a substantial range of couplings, the RFOT may, to all intents and purposes, be considered as a continuous thermodynamic transition at $m = 1$.

We now turn our attention to the variation of the order parameter $h'(r = 0)$ as Γ is gradually lowered along the G_2 branch in the single replica limit $m = 1$. The HNC predictions are shown in Fig. 5. As Γ decreases to values below the RFOT coupling Γ_{cr}=2.12, the "ideal" glass phase becomes metastable relative to the L phase. At $\Gamma = 2.057$, a discontinuous jump of $h'(r = 0)$ is observed towards the G_1 branch, while at $\Gamma < 2.057$, the order parameter drops to its L phase value $h'(r = 0) = 0$. No "glassy" solutions of the HNC equations exist below this coupling, which coincides with the dynamical transition point obtained by Franz et al. (cf. Table I of ref. [20]) for the same model, using a more elaborate approach.

V. EXPLORING THE RANGE $m < 1$

The HNC calculations for a continuously varying number m of replicas can be extended to $m < 1$. In the work of Mézard and Parisi [19,24], the motivation of such an extension is to bypass the RFOT and have access to the free energy of the "ideal" glass phase at temperatures T below the critical temperature T_{cr} via an auxiliary "molecular liquid" phase. Although our own HNC-based search protocol does not require this subtle detour, we explore the range

TABLE I. Γ_{jump}: Γ at which the order parameters $h'(r = 0)$ and Q jump from values corresponding to the G_1 phase to those corresponding to the G_2 phase. Γ_{cr} is the critical Γ at which the free energies of the L and G_2 phases intersect.

m	Γ_{jump}	Γ_{cr}
2.00	1.577	1.650
1.95	1.609	1.675
1.85	1.666	1.734
1.50	1.839	1.910
1.40	1.884	1.955
1.30	1.935	2.060
1.20	1.972	2.044
1.10	2.020	2.088

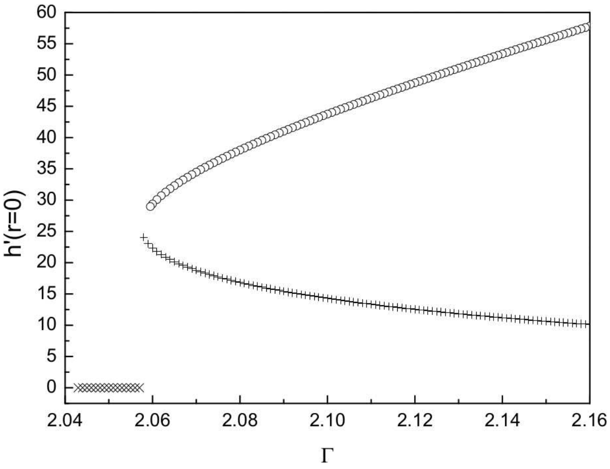

FIG. 5. Variation of the order parameter $h'(r=0)$ with Γ within L (times), G_1 (crosses) and G_2 (circles) phases in the limit $m=1$: succession of two jumps.

$m<1$ to make contact with the Mézard-Parisi predictions. To that purpose we examine the variation of the order parameters $h'(r=0)$ and Q for five values of m, namely 0.75, 0.80, 0.85, 0.90 and 0.95. Independently of m, the order parameters stay close to their L values $h'(r=0)=0$ and $Q=Q_r$ upon increasing Γ until they undergo a discontinuous jump at $\Gamma_{dj}=2.175$ in the limit $\epsilon' \longrightarrow 0$, leading to HNC solutions typical of the G_1 phase (i.e. decreasing order parameters upon increasing Γ). Annealing these states yields solutions typical of the G_2 phase. The variation of the order parameter $h'(r=0)$ with Γ is illustrated in Fig. 6 for $m=0.75$.

We next examine the variation of the excess free energy of the G_1 and G_2 phases with m at fixed temperature, i.e. $f_{ex}^{*G_1}(\Gamma;m)$ and $f_{ex}^{*G_2}(\Gamma;m)$. Two examples are shown in Fig. 7. The behaviours of the two branches differ drastically. Starting from an arbitrary state ($m_0<1$,$\Gamma_0>\Gamma_{cr}$), $f_{ex}^{*G_1}(\Gamma_0;m)$ is seen to be a monotonically increasing function of m, while $f_{ex}^{*G_2}(\Gamma_0;m)$ is found to first increase with m before going through a maximum at some $m^*<1$, i.e. $\partial f_{ex}^{*G_2}/\partial m\big|_{m=m^*}=0$. As the coupling Γ_0 of the initial state is lowered, the position m^* of this maximum is shifted to larger m, until reaching $m^*=1$ for $\Gamma_0=2.13$, in close agreement with the critical value $\Gamma_{cr}=2.12$ reported in Section IV upon approaching $m=1$ from above (i.e. from $m>1$), as illustrated in Fig. 4. As expected, and illustrated in Fig. 7, the free energies of the G_1 and G_2 phases are identical for $m=1$. While for any $m>1$ the free energy of the G_1 phase is systematically higher than that of the G_2 phase, the opposite is true for $m<1$ which appears to imply that the G_1 phase is the stable phase in that range. This awkward situation can however be interpreted upon examining the free energy data for non-zero values of the inter-replica coupling ϵ'. For $\epsilon'>0$ coexistence of low and high overlap phases is observed below a critical temperature T_c [24,25]. Plots of ϵ'(Q) as a function of the overlap order parameter Q for fixed temperature $T<T_c$ ($\Gamma>\Gamma_c$) exhibit van der Waals-like loops, as one might expect from

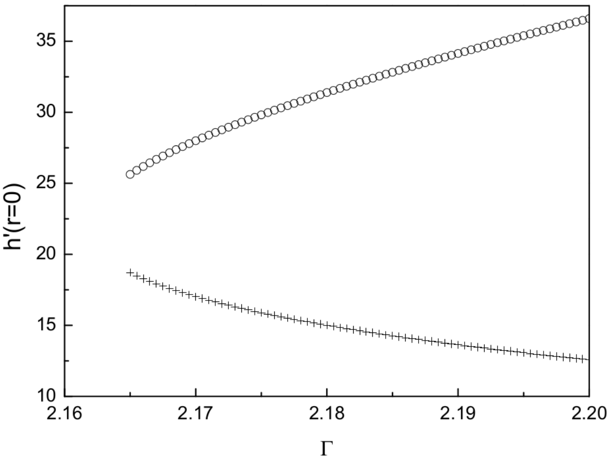

FIG. 6. Variation of the order parameter $h'(r=0)$ with Γ within the G_1 phase (crosses) and within the G_2 phase (circles) for $m=0.75$.

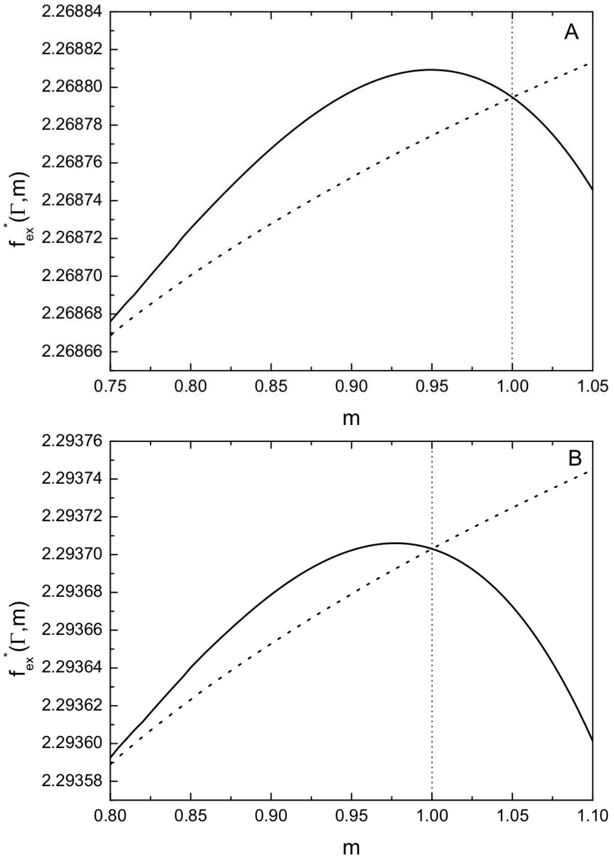

FIG. 7. Variation of the excess free energies of the G_1 (dashed curves) and G_2 (full curves) phases with m for the two couplings $\Gamma = 2.175$ (A) and $\Gamma = 2.15$ (B).

a mean-field theory. These loops involve a low Q branch, associated with the supercooled liquid (L) phase, and a high Q branch, associated with the G_2 "ideal glass" phase, separated by an intermediate branch where ϵ'(Q) is a decreasing function of Q, typical of the G_1 phase. For any $T < T_c$, the Maxwell equal area construction determines the order parameters of the coexisting L and G_2 phases (see e.g. Fig. 4 of [25]). The maximum and minimum of ϵ'(Q) are spinodal points, and spinodal curves are mapped out upon varying $T < T_c$. States inside the spinodal curve, which correspond precisely to the G_1 "phase" are unstable with respect to spinodal decomposition. Note that in the limit $\epsilon' \longrightarrow 0$ T_c=T_{cr}, the RFOT temperature determined in Section IV. Although thermodynamically unstable, the G_1 "phase" might be observable as a transient state, provided the kinetics of spinodal decomposition is sufficiently slow.

VI. CONCLUSION

We have generalized the physically transparent "pedestriant" two-replica analysis of the RFOT to a description based on a continuously varying number m of replicas in the range $2 \geq m > 1$. This extension is easily achieved within the HNC framework, which provides an expression for the free energy, valid for arbitrary m, as well as for the inter-replica pair correlation function $h'(r)$ which allows the calculation of the order parameters $h'(r = 0)$ and Q (the overlap order parameter). For a given m the free energies of the three competing phases L, G_1 and G_2, as identified by the values of the two order parameters, are calculated as functions of the coupling Γ for a supercooled system of particles interacting via the pair potential (1). The critical coupling Γ_{cr} at the RFOT is found to depend sensitively on m; as m decreases, Γ_{cr} increases from $\Gamma_{cr} = 1.65$ for $m = 2$ to $\Gamma_{cr} = 2.12$ in the limit $m \longrightarrow 1$. The question naturally arises of the origin of this significant variation. Is it associated with the replica methodology itself, as applied to structural glasses, or is it due to limitations of the HNC approximation? The latter is well known to be thermodynamically inconsistent, e.g. the virial and compressibility routes lead to significantly different equations of state, particularly in the strong coupling regime [15]. This deficiency can in principle be overcome, by switching to a self-consistent closure relation instead of the HNC closure (6). In our earlier work with $m = 2$, we showed that self-consistent closure relations lead to substantially different estimate of Γ_{cr}, at the cost of a much larger numerical effort, since the HNC free energy expression (7) does not carry over to more advanced closures, and must be replaced by tedious thermodynamic integration protocols [16, 17]. Calculations along these lines, for continuously varying values of m, will be the object of future work. The gradual decrease of T^*_{cr} (increase of Γ_{cr}) with m, predicted by the HNC approximation leads to an additional inconsistency, since for $m \leq 1.84$, T^*_{cr} drops below the m-independent Kauzmann temperature $T^*_K = 0.108$ ($\Gamma_K = 1.74$), which is the temperature at which the configurational entropy (or complexity) of the supercooled liquid vanishes [16]. Such a "cross-over" is of course unphysical, since the RFOT is meant to preempt the "Kauzmann catastrophe", i.e. a negative complexity! Again, future work based on more advanced closure relations should be able to resolve this contradiction.

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