First Principle Computation of Random Pinning Glass Transition, Glass Cooperative Length-Scales and Numerical Comparisons*

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As a guideline for experimental tests of the ideal glass transition (Random Pinning Glass Transition, RPGT) that shall be induced in a system by randomly pinning particles, we performed first-principle computations within the Hypernetted chain approximation and numerical simulations of a Hard Sphere model of glass-former. We obtain confirmation of the expected enhancement of glassy behaviour under the procedure of random pinning, which consists in freezing a fraction c of randomly chosen particles in the positions they have in an equilibrium configuration. We present the analytical phase diagram as a function of c and of the packing fraction ϕ , showing a line of RPGT ending in a critical point. We also obtain first microscopic results on cooperative length-scales characterizing medium-range amorphous order in Hard Spere glasses and indirect quantitative information on a key thermodynamic quantity defined in proximity of ideal glass transitions, the amorphous surface tension. Finally, we present numerical results of pair correlation functions able to differentiate the liquid and the glass phases, as predicted by the analytic computations.

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A common feature of liquids deep below the melting point (supercooled) is the remarkably steep increase of relaxation time, until the system falls out of equilibrium at a conventional temperature $T_{\rm G}$. Since decades, it is present in the literature the claim of the possible presence of a phase transition^{1,2}, the ideal glass transition (IGT), underlying the dynamical arrest and located at a lower temperature $T_{\rm K}$. Despite the first formulation of a consistent phenomenological thermodynamic picture³ this intuition remained at length debated for the lack of other indicators of the imminent thermodynamic singularity. In this context, other theoretical perspectives based on dynamic or topological approaches, sometimes excluding the presence of any transition, have been proposed as alternative explanations of the sluggish dynamics^{4,5}.

In recent times, new important results have been obtained in the development of the thermodynamic scenario. We particularly refer on the one hand, to the first definition and measure of a new kind of cooperative length-scale^{6,7} detecting the spatial extent of amorphous order⁸, called point-to-set l_{PS} , and on the other hand, to the development of a field theory description of the IGT in terms of a suitable large deviation function^{9–11} and the introduction of perturbative^{12,13} and non-perturbative^{14,15} fluctuations in this description. In view of a full fledged theory of glass-formers and quantitative predictions of their properties, these advancements have been corroborated by the formulation of a microscopic theory^{16–19} inspired to classical first-principle computations techniques in liquids^{20,21}.

Due to particularly severe critical properties of the IGT,

i.e. an exponential growth of relaxation time and a power law increase of the cooperative length, the revealing of its properties still remains too difficult to be achieved, leaving fundamental doubts on the whole theoretical picture and on the existence of the IGT itself.

With the aim of giving an answer to these fundamental questions one of us recently proposed^{22,23} a procedure to induce in real systems a glass transition of more easy access than the IGT. The idea is as follows: freeze the position of a fraction c of particles of an equilibrated configuration and study the thermodynamics of the remaining free particles. This procedure should allow the observation of a remarkable growth of the relaxation time in the free-particles equilibrium dynamics and the eventual reaching of a glass transition, called Random Pinning Glass Transition (RPGT), as soon as the concentration of frozen particles reaches a critical value c_K . This result is valid in a full range of moderate and deep supercooling leading to the formation of a line of RPGT, $c_K(T)$, ending in the IGT at T_K and in a new glass critical point at high temperature. This suggests that the way for a test of the theory of IGT is open and calls for numerical $^{24-28}$ and experimental studies of the phase diagram of glassformers with a fraction of frozen particles. On the other side, a microscopic first-principles theory of the RPGT is needed to give more detailed predictions on its physics and provide quantitative information useful in the planning stage of experimental and numerical tests.

In this rapid communication we report the results of a first-principle computation of the RPGT scenario following the method for a quantitative approach to

IGT proposed in Ref. 16: an extension to glasses of the Hypernetted Chain (HNC) approximation of classical theory of simple liquids. Moreover, we report numerical simulation results to be compared with analytic predictions. As glass-former model we chose an Hard Sphere system in 3 dimensions, for which a microscopic thermodynamic theory has already been developed¹⁹ in the unconstrained case. In HS models, particles interact through the usual HS potential $\psi(r) = 0$ for $r \geq D_{ij}$ and ∞ for $r < D_{ij}$, where D_{ij} is the sum of the radii $R_{i,j}$ of particles i and j. HNC computations refer to a monodisperse HSs system. The control parameter ruling the approach to IGT is the density, $\rho = N/V$ for a system with N particles in a volume V, or the packing fraction $\phi = 4\pi R^3 \rho/3$: the fraction of volume occupied by the particles. To prevent crystallization, numerical simulations have instead be realised for a 50:50 binary mixture of N = 250 type A and B spheres of radius $R_B = 1.4 R_A$ and packing fraction $\phi = 2\pi (R_A^3 + R_B^3) \rho/3$. As anticipated in the introduction, the emergence of medium range fluctuations of amorphous order in proximity of the IGT is a long standing open issue. Despite many efforts^{6,14,29,30}, quantitative information on spatial correlation of glass order remained out of reach of the HNC and of other first principles approaches to realistic glass-formers. The present computation, combining together the HNC analytic approach with the pinning particle procedure, at last overcomes this limitation obtaining first quantitative predictions on diverging length-scales of the unconstrained system and even some indirect results on the amorhpous surface tension, a thermodynamic quantity expected to play a major role in the vicinity of the IGT.

According to the Random First Order Transition (RFOT) theory, above a dynamical crossover density ϕ_d (or below T_d), thermodynamics of dense granular systems (or deeply supercooled liquids) starts to be dominated by a large number of particularly stable amorphous configurations corresponding to specific local rearrangements. A non-zero entropy of the $\mathcal N$ stable amorphous configurations can be defined as $S_c = \lim_{N \to \infty} \log(N)/N \neq 0$ and is called configurational entropy. Further increasing the density (or supercooling) the entropy of stable amorphous configurations decreases and vanishes at finite concentration (or temperature) leading in the RFOT theory to the occurrence at ϕ_K (or T_K) of the IGT, a singularity of the thermodynamic entropy (or free-energy) potential. The classical theory of liquids based on diagrammatic expansion of the Morita-Hiroike (M-H) potential^{20,21} has been adapted to capture the effects of this multi-state scenario and the occurrence of the IGT. Among other approximation schemes adopted to compute pair correlation functions, HNC corresponds to a stationary point of a truncated M-H potential where two-line irreducible diagrams have been neglected 21 .

A fundamental step in the formulation of a HNC theory of glass-formers is the introduction of a number of copies (or replicas) of the system and the study of pair correlation functions between particles of different replicas. In practice a system is considered composed by a mixture of particles from different m copies of the original glass-former with positions given by the mN vectors in 3 dimensions $\{x_i^a\}$, where $i \in [1,N]$ is the particle index and $a \in [1,m]$ the replica index. Particles of the same replica interact through the usual pair-potential $\psi(r)$ of the chosen model glass-former, while particles from different replicas do not see each other.

For m=1 the problem becomes a standard HNC liquid computation in terms of usual pair correlation function $g(x,y) = V^2/N^2 \sum_{i \neq j} \langle \delta(x-x_i^a) \delta(y-y_j^a) \rangle$. A pair correlation function between particles of different replicas also appears in the general case: $\widetilde{g}(x,y) = V^2/N^2 \sum_{i,j} \langle \delta(x-x_i^a) \delta(y-y_j^b) \rangle$ with $a \neq b$. Thermodynamic potentials of the glass-former as a function of ϕ (or T) can be computed using the M-H potential of replicated system $S_m^{\text{M-H}}$. The entropy of glass-former 31 is $s = \partial S_m^{\text{M-H}}/\partial m \big|_{m=1}$ and the configurational entropy 31 is $S_c = -m^2 \partial [m^{-1} S_m^{\text{M-H}}]/\partial m \big|_{m=1}$. In the liquid phase, pair correlation functions among different replication functions among different replication functions are standard HNC liquid phase, pair correlation functions among different replication functions are standard formula for the standard functions are standard functions.

In the liquid phase, pair correlation functions among different replicas are trivially equal to one, indicating that replicas are always completely independent. As soon as thermodynamics starts to be dominated by particularly stable configurations, a metastable solution (a new stationary point of the M-H truncated potential) with a non trivial $\widetilde{g}(x,y)$ structure appears. Still, particles of different replicas do not directly interact, but the glassy multistate structure forces different copies of the system to lie in the same stable configuration and originates effective inter-replica couplings.

To study RPGT, we deal with a Hard Sphere system where a fraction c of particles are frozen in an equilibrium reference configuration. We then study the thermodynamics of the remaining free particles replicated m times to probe the formation of a glassy multi-state structure. Finally we average over the equilibrium configurations of frozen particles. In practice, to realize this construction without breaking translational invariance, we will consider a mixture of N(1-c) particles replicated m times (m species), such that particles interact only within the same specie $\psi_{a,a}(r) = \psi(r)$, and $\psi_{a,b}(r) = 0$, and an additional specie (the 0th one) of Nc particles that interact with all particles in the system: $\psi_{0,a}(r) = \psi(r)$. The m copies of N(1-c) particles will hence freely reorganize in presence of the same (pinned) equilibrium template provided by the Nc non-replicated particles³².

The entropy of this mixture of m+1 species can be expressed as prescribed by the M-H potential for mixtures (see Ref.19–21) in terms of densities ρ_{α} with $\alpha \in [0,m]$, of pair correlation functions $g_{\alpha\beta}(x,y)$ and of the Fourier Transform (FT) of $h_{\alpha\beta}(x,y) = g_{\alpha\beta}(x,y) - 1$. These functions are determined by the HNC equations

$$\log g_{\alpha\beta}(x,y) + \psi_{\alpha\beta}(x,y) = h_{\alpha\beta}(x,y) - c_{\alpha\beta}(x,y) \quad (1)$$

and by the Ornstein-Zwernicke closures

$$h_{\alpha\beta}(x,y) = c_{\alpha\beta}(x,y) + \sum_{\gamma} \int dw h_{\alpha\gamma}(x,w) \rho_{\gamma} c_{\gamma\beta}(w,y) .$$
(2)

For m=1, assuming symmetry among the m copies of the free particles, these sets of equations simplify and as in the unconstrained case only two different pair correlation functions, g(x,y) and $\tilde{g}(x,y)$, appear:

$$\log g(x, y) + \psi(x, y) = h(x, y) - c(x, y) \tag{3}$$

$$\log \widetilde{g}(x,y) = \widetilde{h}(x,y) - \widetilde{c}(x,y) , \qquad (4)$$

with

$$h(x,y) - c(x,y) = \rho \int dw h(x,w)c(w,y)$$
 (5)

$$\widetilde{h}(x,y) - \widetilde{c}(x,y) = h(x,y) - c(x,y) + \tag{6}$$

$$-(1-c)\rho\int dw[h(x,w)-\widetilde{h}(x,w)][c(w,y)-\widetilde{c}(w,y)]\ .$$

The first ones imply a solution g(r) independent from $\widetilde{g}(r)$ and identical to the simple liquid solution³³. The second ones admit two different solutions, $\widetilde{g}_{L}(r)$ and $\widetilde{g}_{\rm G}(r)$, both present in some ranges of the control parameters ϕ (or T) and c. These two solutions encode respectively the liquid (L) low correlations between particles of different replicas induced by the presence of a fraction of Nc particles, and the glass (G) high correlations effectively generated by the multi-state structure. In the M-H entropy expression we deal with matrices of pair correlation functions $\sqrt{\rho_{\alpha}\rho_{\beta}}g_{\alpha\beta}$ with only four different elements that, assuming translational and rotational invariance, is: $\rho_0 g_{00} = c \rho g(r)$, if $\alpha \neq 0$ $\sqrt{\rho_0 \rho_\alpha} g_{0\alpha} =$ $\sqrt{\rho_0 \rho_\alpha} g_{\alpha 0} = \sqrt{c(1-c)} \rho g(r)$, and $\rho_\alpha g_{\alpha \alpha} = (1-c) \rho g(r)$, and if also $\beta \neq 0$ $\sqrt{\rho_{\alpha}\rho_{\beta}}g_{\alpha\beta} = \sqrt{\rho_{\alpha}\rho_{\beta}}g_{\beta\alpha} = (1-c)\rho \widetilde{g}(r)$. Hence the entropy of glass-former, obtained from the M-H potential of replicated system $S_m^{\text{M-H}}$, reads

$$s[\phi, c; g, \widetilde{g}] = -\frac{\rho}{2} \int 4\pi r^2 dr (1-c) \left[(1+c)g(r)(\log(g(r)) - 1) + (1-c)\widetilde{g}(r)(\log(\widetilde{g}(r)) - 1) + (1+c)\psi(r)g(r) + 2 \right] + (7) + \frac{1}{2\rho} \int_q (1-c) \left[-(1+c)\rho h + c\rho\widetilde{h} + \frac{1}{2}(1+c)\rho^2 h^2 + \frac{1}{2}(1-c)\rho^2\widetilde{h}^2 + \rho\frac{ch + (1-c)\widetilde{h}}{1+\rho h} + \frac{\log\left(1 + (1-c)\rho(h - \widetilde{h})\right)}{1-c} \right],$$

where h and \widetilde{h} are the FTs of h(x) and $\widetilde{h}(x)$, and \int_q represents integration in the 3d momentum space³⁴.

Through numerical iterative solution³⁵ of Eqs.(3-6) we can obtain: $\widetilde{q}_{L}(r)$ using $\widetilde{q}(r) = 1$ as initial condition, and $\widetilde{q}_{G}(r)$ when the initial condition is the non-trivial $\widetilde{q}(r)$ solution of unconstrained systems. We can also compute the entropy and hence the stability of the corresponding phases. For $\phi \in (\phi_d, \phi_K)$ and c = 0 the entropy of L is larger than the entropy of G and the L solution is stable. When c increases, for fixed ϕ , the difference in entropy between the two phases decreases, vanishes at $c_K(\phi)$, and eventually changes sign for $c > c_K(\phi)$ where the G solution becomes stable (see Fig.1). Finally, the Lphase disappears beyond the spinodal line³⁶ $c_s(\phi)$. When particles are pinned from equilibrium configurations, the entropy (or more in general the free-energy) mismatch between the L and G phases coincides with the configurational entropy of the constrained system (see appendices of Ref.23). Hence, the transition occurring at $c_K(\phi)$ is an actual entropy-vanishing transition, the RPGT, with similar features to the IGT²². For $\phi < \phi_d$, a second spinodal, $c_d(\phi)$, of the G phase appears and the RPGT line continues in this low concentration regime, indicating that in a HS model the glass solution can be generated by pinning particles even if it was completely absent in the unconstrained system. When ϕ decreases, the two spinodals $c_d(\phi)$ and $c_s(\phi)$ slowly approach the transition line $c_K(\phi)$ and the two solutions $\widetilde{g}_L(r)$ and $\widetilde{g}_G(r)$ computed at $c_K(\phi)$ approach each other (see inset of Fig.1). The three lines meet in a critical point 12,13,15,22 $\{c^*,\phi^*\}$, where $\widetilde{g}_L(r)$ and $\widetilde{g}_G(r)$ eventually merge. For lower packing fraction, random pinning only induces trivial pair correlation $\widetilde{g}_L(r)$ among particles.

The HNC study of randomly pinned systems allows us to obtain the first microscopic results on cooperative lengthscales of non-trivial glass fluctuations and an indirect evaluation of a key thermodynamic quantity of RFOT: the free-energy cost of the matching between different amorphous stable configurations, a.k.a. the amorphous surface tension. From $c_K(\phi)$ the glass phase is stable: the system is able to spontaneously reconstruct the template configuration starting from the local constraints imposed by pinned particles. This reveals in the template configuration the presence of a subtle medium-range correlation that extends over length-scales smaller than the typical distance ξ between pairs of pinned particles at criticality: $\xi(\phi) \sim c_K^{-1/d}(\phi)$, where d=3 is the dimensionality of the system. In quite perfect agreement with phenomenological scaling arguments, we obtain for $\xi(\phi)$ vs $\phi_k - \phi$ a simple inverse cubic square behavior (see Fig.2), ruled by the almost linear vanishing of $S_c(\phi) \sim \phi_K - \phi$, except for mild deviations in the pre-asymptotic range. Moreover, we notice that pinning a finite fraction of particles leads 22 to a decrease of the configurational entropy of the orig-

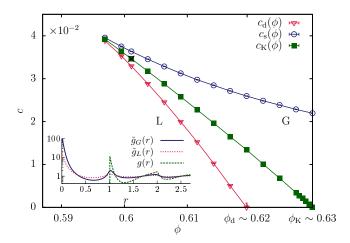


FIG. 1. Phase diagram of the randomly pinned Hard Sphere model in HNC approximations. A RPGT line is expected to begin in the IGT of unconstrained system and end in a critical glass transition point $\{c^*, \phi^*\}$. Two spinodal lines of the liquid L $(c_s(\phi))$ and ideal glass G $(c_d(\phi))$ phases are also reported. The inset shows the g(r), $\tilde{g}_L(r)$ and $\tilde{g}_G(r)$ in correspondence of the cross in the phase diagram $(\phi = 0.601) > \phi^*$ and $c_K(\phi = 0.601)$.

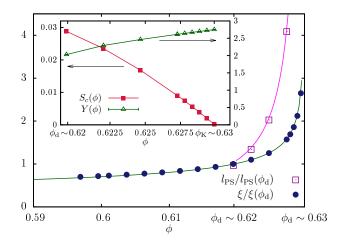


FIG. 2. Growth of non-trivial correlation length-scales of amorphous order. Small corrections to the power law behavior with exponent -1/3 are present in the pre-asymptotic growth of ξ . The much faster increase of l_{PS} follows a power law divergence with exponent -1. Inset: configurational entropy and an indirect result on the amorphous surface tension of the unpinned system.

inal system $S_c(\phi)$ rougly proportional to c, when c is small. Hence the configurational entropy for the pinned system is $S_c^P(\phi,c) \simeq S_c(\phi) - cY(\phi)$ and $Y(\phi)$ is a microscopic configurational entropy loss due to the locally imposed constraint, a quantity complementary²² to the amorphous surface tension: the interface free-energy cost between typical amorphous configurations. We can easily compute its value from a linear fit of the configurational entropy decrease due to small pinning. We consistently find that Y is only defined where $S_c(\phi)$ exists, hence

above ϕ_d , and it moderately increases when the IGT is approached^{37,38}, as it is shown in the inset of Fig.2. Finally, we can compute a second cooperative length scale, called point-to-set, l_{PS} . This length was initially operatively defined by using an alternative pinning procedure where all the particles are pinned except those in a cavity of size l. Phenomenological arguments on that construction gave as a result³⁹ $l_{PS} \sim Y(\phi)/S_c(\phi) \sim \xi(\phi)^d$ and we can compute it having obtained $S_c(\phi)$ and $Y(\phi)$ from the present HNC computation. In Fig.2 the two cooperative length scales rescaled to 1 at ϕ_d are compared showing a much faster growth of l_{PS} , highlighting the difference between the two lengths, and indicating the convenience of the point-to-set procedure to reveal growth of amorphous order in real systems. Note that this result warns about actual important differences between apparently analogous procedures to detect cooperative length scales⁴⁰.

In this last part, we study the validity of the HNC picture described above via numerical simulations of HSs. Despite the advantages of detecting a transition at ϕ well

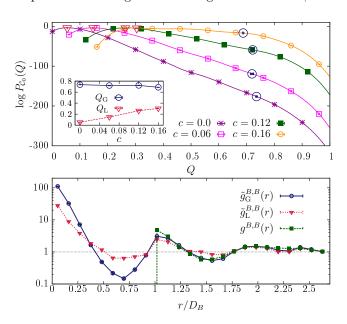


FIG. 3. (Top) Logarithm of $P_{C_0}(Q)$ for different values of c. The big triangles represent $Q_{\rm L}$ and $Q_{\rm G}$ and they are plotted as function of c in the inset. (Bottom) The pair correlation functions (for the B particles) at the two maxima for c = 0.16.

below $\phi_{\rm K}$, probing numerically the existence of an ideal transition line $c_{\rm K}(\phi)$ in equilibrium is still very hard. Indeed, dynamics at $\phi\gtrsim\phi_{\rm d}$ is already very slow, and from there, characteristic times grow very fast as $c_{\rm K}(\phi)$ is approached. In this study we only attempt to equilibrate in the liquid part of the phase diagram in Fig.1. Still we can to obtain an indication of $c_{\rm K}(\phi)$ by studying the relative weight between the (metastable) glass phase G and the (stable) liquid phase L.

As in the analytical calculations, we chose a reference configuration C_0 from a well equilibrated system at $\phi = 0.58$, and frozen (pin) the positions of cN particles (c = 0, 0.06, 0.12 and 0.16). Since here we just want

to show qualitatively that the random pinning does enhance the glassy behavior, we will only present results for a single reference configuration C_0 , at which we are able to thermalize up to high values of c (a complete numerical study of the transition will be presented elsewhere⁴¹). To tackle with thermalization issues, we apply the reversible Event Chain Monte Carlo (ECMC) algorithm 42,43 (with a slight variation to account for the cNimmobile variables⁴⁴), which represents a gain of a factor 10 in times with respect to standard Monte Carlo (MC) moves. In addition to that, we used the tethered MC method⁴⁵ to quantify the relative weight between the L and G phases without waiting for the occurrence of spontaneous activated events during the dynamics. The tethered strategy⁴⁵ relies on independent simulations at fixed values of the order parameter, in this case the overlap $Q(\mathcal{C}, \mathcal{C}_0)$ between the running configuration \mathcal{C} and the reference one \mathcal{C}_0 (the overlap is defined as $Q(\mathcal{C}, \mathcal{C}_0) = \frac{1}{N} \sum_{i}^{N_b} (n_{i,A}^{\mathcal{C}} n_{i,A}^{\mathcal{C}_0} + n_{i,B}^{\mathcal{C}_0} n_{i,B}^{\mathcal{C}_0})$ where n_i are the occupation variables of the N_b small boxes of size $\ell \lesssim 1/\sqrt{3}D_{A,A}$: $n_i = 1$ if occupied 0 if not). One can recover the full probability distribution function, $P_{\mathcal{C}_0}(Q) = \langle \delta(Q - Q(\mathcal{C}, \mathcal{C}_0)) \rangle_{\mathcal{C}_0}$, being $\langle \cdot \rangle_{\mathcal{C}_0}$ the equilibrium average being C_0 fixed, via a thermodynamic integration on the mesh of Q simulation-points¹¹. We plot the logarithm of $P_{\mathcal{C}_0}(Q)$ in the top panel of Fig.3 for several values of c. $P_{\mathcal{C}_0}(Q)$ always has a maximum at $Q_{\mathrm{L}}(c)$, and develops an elbow at high values of Q that becomes more pronounced as c is increased: a clear indication of the glassy enhancement upon pinning particles. Since the second glass maximum is never properly formed, as it is expected beyond mean-field, we identify the $Q_{\rm G}(c)$ with the position at which the high-Q maximum would

be if an external field ϵ coupled to the overlap were introduced to make $Q_{\rm L}^{\epsilon}(c)$ and $Q_{\rm G}^{\epsilon}(c)$ equally probable (see Refs. 10,11,46 for technical details). The behavior of $Q_{\rm L}$ and $Q_{\rm G}$ with c is shown in the inset of Fig.3. The IGT is given by the point at which the two maxima are equally probable, which occurs here $c_{\rm K} \gtrsim 0.16$. We can study the corresponding $\widetilde{g}_{\rm L}(r)$ and $\widetilde{g}_{\rm G}(r)$ at these two maxima (see bottom panel of Fig.3). Now $\widetilde{g}_{L,G}(r)$ is the pair-correlation function between particles in configuration C_0 and in $\mathcal C$ when $Q=Q_{L,G}$. In a bidisperse system, one can compute three different $\widetilde{g}(r)$: $\widetilde{g}_{L,G}^{AA}(r)$, $\widetilde{g}_{L,G}^{AB}(r)$ and $\widetilde{g}_{L,G}^{BB}(r)$. We just show the $\widetilde{g}_{L,G}^{BB}(r)$ computed with the small particles, since $\widetilde{g}_{L,G}^{AA}(r)$ displays the same qualitative behavior. $\widetilde{g}_{L}^{BB}(r)$ and $\widetilde{g}_{G}^{BB}(r)$ show two distinct behaviours but they appear to be at the edge of merging as it is expected in proximity of the end of the RPGT line, see inset in Fig.1.

We presented the microscopic results of first principle HNC computations and of numerical simulations in a Hard Sphere model glass-former with a fraction of frozen particles. The analytical and numerical results confirm expectations on the existence of a new kind of glass transition, called RPGT, induced by pinning particles and provide microscopic information on its occurrence in the $\phi - c$ phase diagram. First microscopic results on nontrivial static cooperative length scales of glass order and on the amorphous surface tension also derive from the HNC computation. Finally first numerical results on $\widetilde{g}_{L,G}(r)$ in a bidisperse HS model simulated with an optimized Monte-Carlo dynamics have been presented and confirm the analytic predictions on the appearance of a glass phase when pinning is increased and on the features of $\widetilde{g}_G(r)$ compared to the trivial $\widetilde{g}_L(r)$.

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