Computer simulations of the Gardner transition in structural glasses

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I. CONNECTIONS BETWEEN GARDNER AND SPIN-GLASS TRANSITIONS

The exact mean-field theory for the simplest glass-forming system - the dense assembly of hard spheres in the large dimensional limit - predicts the existence of a Gardner phase $[1, 2]$. This transition is characterized by full replica symmetry breaking (RSB) that implies two fascinating physical consequences. (i) A hierarchical free-energy landscape, i.e., the thermal fluctuations are organized hierarchically, meaning that configurations are grouped into meta-basins that are further grouped into meta-meta basins, ... (ii) The marginal stability, i.e., the system responds sensitively to infinitesimal perturbations. Here we discuss recent results of numerical simulations to examine these mean-field predictions in physical dimensions.

From the viewpoint of RSB, the Gardner transition in structural glasses belongs to the same full RSB universality class of the spin-glass transition (see Fig. 1(A)). This theoretical ground motivates us to borrow ideas from the extensive research on spin-glasses to study the Gardner transition. To this end, it is useful to review firstly some of the essential results obtained in spin-glass experiments and simulations.

The RSB solution immediately implies a hierarchy of linear responses through the fluctuation-dissipation relation $[3]$. One expects short-time, intermediate-time, and long-time linear responses associated with thermal fluctuations inside basins, meta-basins, and meta-meta-basins. A remarkable consequence is the “anomaly” that gives a natural explanation for the protocol-dependent linear responses observed experimentally $[4]$. In one protocol called field cooling (FC), one measures the magnetization $m_{\text{FC}}$ of a spin-glass under cooling from a high temperature $T_{\text{max}}$ down to a low temperature $T_{\text{min}}$ below the spin-glass transition temperature $T_{\text{SG}}$ in the presence of a weak external magnetic field $\delta h$; in the other protocol called zero field cooling (ZFC), one cools the spin-glass from $T_{\text{max}}$ down to $T_{\text{min}}$ without the field ($h = 0$), then switches on the magnetic field $\delta h$ and measures the magnetization $m_{\text{ZFC}}$ under heating the spin-glass back to $T_{\text{max}}$ (see Fig. 2(B)). The two susceptibilities $\chi_{\text{FC}} = m_{\text{FC}}/\delta h$ and $\chi_{\text{ZFC}} = m_{\text{ZFC}}/\delta h$ are the same above $T_{\text{SG}}$, but different ($\chi_{\text{FC}} > \chi_{\text{ZFC}}$) below (see Fig. 2(A)). The fact that $\chi_{\text{FC}}$ and $\chi_{\text{ZFC}}$ are different in the spin-glass phase is referred to as an “anomaly”, because the susceptibility is protocol-independent in standard magnetic systems. The RSB theory gives $\chi_{\text{FC}} - \chi_{\text{ZFC}} = \beta \left[ \int_0^1 dq P(q) q - q_{\text{EA}} \right]$, with $\beta$ the inverse temperature. Here $q_{\text{EA}}$ is the Edwards-Anderson (EA) order parameter $[5]$ representing the strength of the thermal fluctuation within lowest-level basins, while $\int_0^1 dq P(q) q$ represents the integral of thermal fluctuations coming from all levels in the hierarchy. In the replica symmetric (RS) solution, $P(q) = \delta(q - q_{\text{EA}})$, so that the anomaly vanishes.
The anomaly is known to be not a transient but a long-time effect, as demonstrated by a series of experiments that reveal aging effects in spin-glasses [7–9]. To study the dynamical effects, one can generalize the ZFC protocol by introducing a waiting time $t_w$ before switching on the magnetic field, and a measurement time $\tau$ elapsed in the presence of the field. By increasing $\tau$, $m_{ZFC}(\tau, t_w)$ increases passing through $m_{ZFC}$ and heads toward $m_{FC}$ (see Fig. 2(B)). However, $m_{ZFC}(\tau, t_w)$ does not reach $m_{FC}$ within finite time, and its time evolution as a function of $\tau$ slows down with increasing waiting time $t_w$, manifested by scaling laws depending on $\tau/t_w$. These experimental observations are significant because they reveal the out-of-equilibrium nature of spin-glasses. To describe the aging effects and the anomaly from a purely dynamical point of view, a dynamical mean-field theory on spin-glass models is developed [10, 11]. The dynamical theory relates RSB to the notion of effective temperature that characterizes out-of-equilibrium glassy dynamics [12, 13]. The numerical evidence of effective temperature [14] and non-zero anomaly in the long-time limit [15] has been indicated by detailed simulations of finite-dimensional spin-glass models.

The marginal stability of the spin-glass phase may account for various complex non-linear responses, such as the effect of static chaos with respect to an infinitesimal change of temperature, or avalanches with respect to an infinitesimal change of magnetic field. Indeed the equilibrium spin configurations at large length scales are completely reshuffled by infinitesimal perturbations, which is predicted first by the droplet theory [16–18], and later by theories based on RSB [19–26]. The rejuvenation-memory effects observed experimentally [27] may be related to such non-linear responses [28, 29].

Once one is aware of the correspondence between spin and structural glasses (see Fig. 1(A)), it is natural to use strategies inherited from spin-glass studies to explore the physics of Gardner phase in structural glasses. For example, in the ZFC/FC protocols, the role of the magnetic field $h$ for spin-glasses can be replaced by the shear strain $\gamma$ for structural glasses (see Fig. 1(B)), which only changes the boundary condition but not the thermodynamic properties of the bulk. Indeed, the replica theory of structural glasses predicts a hierarchy of shear moduli reflecting RSB [30–32]. By adapting the methods developed in spin-glasses, one can examine the aging effects, the protocol-dependent linear responses, and the non-linear responses such as avalanches, in structural glasses with respect to shear deformations. Our discussion focuses on one of the simplest models of structural glasses in three dimensions, hard spheres, where the (reduced) pressure $p$ (or the volume fraction $\varphi$) plays the role of temperature $T$. According to the replica theory [1] that is exact in the large dimensional limit, a Gardner transition occurs in hard spheres under both compression and shear, which is examined by simulations at three-dimensions in the following sections. The dynamical mean-field theory for the hard-sphere glass has also been set up [33–36], but a detailed theoretical analysis of the out-of-equilibrium dynamics remains challenging. Nonetheless, the analogy to the spin-glass problem outlined above allows us to infer the implications of RSB on the dynamics of hard spheres.
II. GARDNER TRANSITION UNDER COMPRESSION

A. Preparation of ultra-stable glasses

To study the Gardner transition, we must prepare a glass at first. Experimentally, glasses are obtained by a slow thermal or compression annealing, the rate of which determines the location of the glass transition. It is found that a detailed numerical analysis of the Gardner transition requires the preparation of extremely well-relaxed glasses (corresponding to structural relaxation timescales challenging to simulate in standard algorithms), in order to study vibrational motions of particles without interference from diffusion. Such ultra-stable glasses can be numerically generated by applying a swap Monte-Carlo scheme [37, 38] to a simple glass-forming model – a polydisperse mixture of $N$ hard spheres [39].

The annealing procedure contains two steps [39]. First, one produces equilibrated liquid configurations at various densities $\varphi_g$ with the help of the swap algorithm. Second, starting from these liquid configurations, one switches to standard molecular dynamics simulations [40] during which the system is compressed out of equilibrium up to target densities $\varphi > \varphi_g$. In order to obtain thermal and disorder averaging, this procedure is repeated over many samples, each corresponding to different initial equilibrium configurations at $\varphi_g$, and over many independent quench realizations for each sample. The independent realizations of the same sample have identical particle positions at $\varphi_g$, but are assigned to different initial velocities drawn from the Maxwell–Boltzmann distribution.

The above numerical protocol is analogous to thermal annealing with different cooling rates, which results in different glass transition temperatures. Each glass transition density $\varphi_g$ selects a particular glass state. The value of $\varphi_g$ ranges from the mode-coupling theory (MCT) density (or the dynamical glass transition density) $\varphi_d$, at which the liquid relaxation is slow but affected by activated $\alpha$-processes, to $\varphi_g \gg \varphi_d$, where particle diffusion and vibrations are fully separated. For sufficiently large $\varphi_g$, the $\alpha$-relaxation time becomes larger than the simulation time by many orders of magnitude; one thus obtains unimpeded access to the dynamics within the glass state, i.e., the $\beta$-relaxation processes [41].

The liquid equation of state (EOS) for the reduced pressure $p = \beta P/\rho$ of the model, where $\rho$ is the number density, and $P$ the system pressure, can be well described by the Carnahan-Starling (CS) equation [42]. The dynamical glass transition density $\varphi_d = 0.594(1)$ was estimated following the strategy in Ref. [43]. Note that the dynamical glass transition is only rigorous in large dimensions; it becomes a dynamical crossover in three dimensions (see Chapter 16 for a detailed discussion). The non-equilibrium glass EOSs associated with compression terminate at inherent states (where $p \rightarrow \infty$) that correspond to, for hard spheres, jammed configurations at $\varphi_j$, and can be captured by a free-volume scaling form, $p_{\text{glass}}(\varphi) \sim (\varphi_j - \varphi)$ [44]. Figure 4 presents the phase diagram and EOSs of the model.

Along each glass EOS of a given $\varphi_g$, a corresponding Gardner transition may exist at density $\varphi_G$ (or pressure $p_G$), as predicted by the mean-field theory. For $\varphi_g < \varphi < \varphi_G$, the system is in a stable glass phase: each glass state is confined in one of the structureless basins on the free-energy landscape (see Fig. 4(A)), and is stable in response to
small mechanical deformations. On the other hand, the regime $\phi_G < \phi < \phi_J$ corresponds to a marginal glass phase, where each simple glass basin splits into a fractal hierarchy of sub-basins and the glass becomes marginally stable to deformations. The Gardner line and the liquid EOS merge around $\phi_d$, suggesting the mixing of dynamical behavior associated to the Gardner transition and to the glass transition – this is why one needs to focus on ultra-stable glasses in order to explore pure Gardner physics.

B. Key observables and protocols

In the glass state, particles vibrate inside their cages (see the insets of Fig. 3). The first approach to study the Gardner transition is based on the direct analysis of caging order parameters, which quantify the caging properties of particles. In the case of stable glasses, the caging order parameter is defined as, $\Delta_{EA} = \lim_{t \to \infty} \frac{1}{N} \sum_{i=1}^{N} \langle |\vec{r}_i(t) - \vec{r}_i(0)|^2 \rangle$, where $\vec{r}_i(t)$ is the position of particle $i$ at time $t$. The parameter $\Delta_{EA}$, which decreases with the degree of annealing, corresponds to nothing but the EA parameter $q_{EA}$ in spin-glasses.

Similar to the spin-glass transition, the Gardner transition induces the split of basins on the free-energy landscape and aging effects, which suggests that the order parameter must be generalized: one considers (i) the mean-squared displacement (MSD) $\Delta(\tau, t_w)$ and (ii) the distance between pairs of independently quenched configurations $\Delta_{AB}(t)$. Here the MSD is defined as, $\Delta(\tau, t_w) = \frac{1}{N} \sum_{i=1}^{N} \langle |\vec{r}_i(\tau + t_w) - \vec{r}_i(t_w)|^2 \rangle$, averaged over both thermal fluctuations and disorder, at the target $\phi$ reached by compression. A waiting time $t_w$ is introduced in order to explicitly examine the aging effects (the total time $t$ is the sum of the measurement time $\tau$ and $t_w$). On the other hand, $\Delta_{AB}(t) = \frac{1}{N} \sum_{i=1}^{N} \langle |\vec{r}_A^i(t) - \vec{r}_B^i(t)|^2 \rangle$, where the two copies $A$ and $B$ are independent realizations at $\phi$, compressed from the same initial sample at $\phi_g$.

The large-time limits of these quantities have important physical meanings. The EA order parameter is defined as $\Delta_{EA} \equiv \lim_{\tau \to \infty} \lim_{t_w \to \infty} \Delta(\tau, t_w)$. Here the order of time limits is crucial [45]: by reversing the order one can define another parameter, $\Delta_{AB} \equiv \lim_{t_w \to \infty} \lim_{\tau \to \infty} \Delta(\tau, t_w) = \lim_{t \to \infty} \Delta_{AB}(t)$. The RSB is signaled by $\Delta_{AB} > \Delta_{EA}$ (note that $\Delta_{AB} = \Delta_{EA}$ in stable glasses). In other words, the two large-time limits cannot be interchanged in the Gardner phase, meaning that the aging effects become persistent.

In the second approach, one studies the response of hard-sphere glasses against a shear strain $\gamma$, analogous to observing magnetic susceptibilities in spin-glasses. The simple strain $\gamma$ is applied to the $x$-coordinates of all particles $(x_i \to x_i + \gamma z_i)$ after a waiting time $t_w$, under the constant-volume and Lees-Edwards boundary conditions [46]. The strain is increased slowly with a constant shear rate $\dot{\gamma}$, and the reduced shear stress $\sigma = \beta \Sigma / \rho$ is measured, where $\Sigma$
is the stress (for convenience, some data are presented with the unitless stress rescaled by pressure, $\tilde{\sigma} = \sigma/p$).

As in the spin-glass case, one can consider two types of protocols, namely zero field compression (ZFC) and field compression (FC) (see Fig. 2(B)). In the ZFC protocol, one compresses the configuration from $\varphi_p$ to $\varphi$, waits for time $t_w$ before applying a shear strain $\delta\gamma$ instantaneously, and then measures the stress $\sigma_{ZFC}(\tau, t_w)$ as a function of $\tau$. In the FC protocol, one applies $\delta\gamma$ at the initial density $\varphi_0$, and then measures the stress $\sigma_{FC}(t)$ once the configuration is compressed to $\varphi$ ($t$ is reset to zero after compression). Similar to the caging order parameters, two large-time limits can be considered: $\sigma_{ZFC} = \lim_{\tau \to \infty} \lim_{t_w \to \infty} \sigma_{ZFC}(\tau, t_w)$ and $\sigma_{FC} = \lim_{\tau \to \infty} \lim_{t_w \to \infty} \sigma_{ZFC}(\tau, t_w)$.

Theories have demonstrated that the above two approaches (more specifically, the caging order parameters $\Delta$ and the shear moduli $\mu$) are intrinsically related \cite{32}: in the large pressure limit, $\mu_{ZFC} \sim 1/\Delta_{\text{EA}}$ and $\mu_{FC}/p \sim 1/\Delta_{\text{AB}}$. These relationships are the counterpart of the duality between overlapping order parameters and magnetic susceptibilities in spin-glasses.

C. Aging effects

![Figure 4](image)

FIG. 4. Time evolutions of (A) caging order parameters (adapted from \cite{39}) and (B) shear moduli (adapted from \cite{47}), in the Gardner phase of the hard-sphere glass model.

In the Gardner phase ($\varphi > \varphi_G$), aging effects can be observed in both MSD (without shear deformations) and shear responses. Figure 4(A) shows the simulation data of MSD. After a short time $\tau_0 \sim 1$ of ballistic motions, the evolution of $\Delta(\tau, t_w)$, as a function of $\tau$, exhibits a plateau followed by further growth. The switch from the former to the latter happens at longer times with increasing waiting time $t_w$. The height of the short-time plateau gives $\Delta_{\text{EA}} = \Delta(\tau = \tau_0, t_w = 0)$. Figure 4(A) also displays $\Delta_{\text{AB}}(t)$, which is time-independent and should correspond to a long-time plateau of $\Delta(\tau, t_w)$ (this plateau is unfortunately beyond the current simulation time window). The clear separation of the two parameters ($\Delta_{\text{AB}} > \Delta_{\text{EA}}$) is the first numerical evidence of the ergodicity breaking in the Gardner phase \cite{39, 48, 49}.

Figure 4(B) shows the time-dependent (unitless) shear moduli, $\tilde{\mu}(\tau, t_w)$, whose behavior is similar to that of MSD. An important feature is that $\tilde{\mu}_{ZFC}(\tau, t_w)$ exhibits a plateau suggesting the existence of $\tilde{\mu}_{ZFC}$. On the other hand, $\tilde{\mu}_{FC}(t)$ is essentially a constant in time $t$ (for $t > \tau_0$), which shall be denoted as $\tilde{\mu}_{FC}$. In the proper order of large-time limits, one expects that $\tilde{\mu}_{ZFC}(\tau, t_w)$ decays to $\tilde{\mu}_{FC}$, as $\lim_{t_w \to \infty} \lim_{\tau \to \infty} \tilde{\mu}_{ZFC}(\tau, t_w) = \tilde{\mu}_{FC}$, but the convergence becomes slower as $t_w$ increases. Apparently $\tilde{\mu}_{ZFC}$ is larger than $\tilde{\mu}_{FC}$, which parallels $\Delta_{\text{AB}} > \Delta_{\text{EA}}$.

D. Anomalous order parameters and responses

Figure 5 shows the pressure dependence of caging order parameters ($\Delta_{\text{EA}}$ and $\Delta_{\text{AB}}$) and shear moduli ($\tilde{\mu}_{ZFC}$ and $\tilde{\mu}_{FC}$) obtained through the above-mentioned dynamic measurements. One finds that, in the stable glass phase ($p < p_G$), $\Delta_{\text{EA}} = \Delta_{\text{AB}}$ and $\tilde{\mu}_{ZFC} = \tilde{\mu}_{FC}$, while in the Gardner phase ($p > p_G$), $\Delta_{\text{EA}} < \Delta_{\text{AB}}$ and $\tilde{\mu}_{ZFC} > \tilde{\mu}_{FC}$.
large pressure limit, mean-field theories predict that \( \Delta_{EA} \sim p^{-\kappa} \) [50] and \( \mu_{ZFC} \sim p^\kappa \) [32], where \( \kappa = 1.41574 \). The former is verified by three-dimensional simulations in Ref. [50] and the latter by those in Ref. [47] (see Fig. 5(B)). The theories also give large-\( p \) predictions \( \mu_{FC}/p \sim 1/\Delta_{AB} \sim \text{constant} \), which are consistent with the simulation results in Fig. 5.

**FIG. 5.** (A) Bifurcation of caging order parameters \( \Delta_{EA} \) and \( \Delta_{AB} \) around the Gardner transition \( p_G \approx 2.7 \times 10^2 \) (adapted from [39]). (B) Protocol-dependent shear moduli \( \tilde{\mu}_{ZFC} \) and \( \tilde{\mu}_{FC} \) (adapted from [47]). The solid line indicates the scaling \( \mu_{ZFC} \sim p^{1.41574} \) predicted by the mean-field theory [32].

### III. GARDNER TRANSITION UNDER SHEAR

As predicted theoretically [51], a Gardner transition at \( \gamma_G \) could occur under shear, before the glass yields at \( \gamma_Y \). Figure 6(A) shows the stability map of hard sphere glasses under shear and compression/decompression [52, 53]. The Gardner transition and yielding give rise to three types of behavior in a typical cyclic shear test (see Fig. 6(B)).

(i) The stress-strain curve is reversible in the stable glass phase (\( \gamma < \gamma_G \)). (ii) If the shear strain is reversed at a maximum strain \( \gamma_{\text{max}} \) between \( \gamma_G \) and \( \gamma_Y \), a hysteresis loop emerges, which however disappears below \( \gamma_G \). This partial-reversible phenomenon is a manifestation of the hierarchical free-energy landscape consisting of basins within a common meta-basin. The part of the stress-strain curve in the Gardner phase (\( \gamma_G < \gamma < \gamma_Y \)) is jerky due to many small avalanches, reflecting the marginal stability. (iii) If \( \gamma_{\text{max}} > \gamma_Y \), the cycle becomes strongly irreversible, suggesting the destruction of glass meta-basin after yielding.

### IV. DISCUSSION AND OUTLOOK

As a second-order phase transition, the fluctuation of order parameters (or the susceptibility) is expected to diverge at the Gardner transition in the thermodynamic limit. Simulations have shown that the caging susceptibility grows orders of magnitude approaching the Gardner transition [39]. Furthermore, the spatial correlations between local caging order parameters become long-ranged in the Gardner phase, implying the heterogeneity of vibrational dynamics [39, 54]. However, dynamical activations could possibly turn a mean-field thermodynamic phase transition into a crossover in low dimensions. It remains inconclusive whether a sharp Gardner transition survives in three dimensions, although a renormalization group theory based on loop expansions [55] (see Chapter 3 for details) and a machine-learning facilitated finite-size analysis of simulation data [55] seem to suggest so.

The discussion so far has focused on the hard-sphere model. Hard spheres have a well-defined singularity under compression, the jamming transition, where quantities such as pressure and the length scale of mechanical response diverge (see Chapter 19 for a review on the jamming transition). Because the jamming transition lies in the Gardner phase, the full RSB predictions should also apply to the criticality and marginality of jamming, which are quantified by power-law scalings of weak forces, small interparticle gaps [50] and low-frequency vibrational modes [57]. Remarkably,
The numerical results seemingly agree with the mean-field exponents even in low dimensions \cite{50, 58, 59}. The evidence of ultrametricity that characterizes the hierarchical energy landscape, has also been demonstrated numerically in jammed packings in three dimensions \cite{60}.

The Gardner transition seems to emerge as a “precursor” of certain singularities (jamming under compression and yielding under shear) in hard particles. The situation is more complicated in cases without such singularities, e.g., cooling soft spheres under the constant density condition. On the one hand, the mean-field theory universally identifies the existence of Gardner transition in soft spheres \cite{61}, and simulations have reported a rejuvenation-memory effect \cite{62} similar to that found in spin-glasses. On the other hand, however, simulations demonstrate that the Gardner transition could be interfered with by low-dimensional effects such as localized “defects” \cite{63}. Separating the Gardner physics from strong low-dimensional effects in soft spheres remains a challenge in simulations.

Finally, experimental efforts to detect the Gardner transition have shown encouraging progress. The caging order parameter approach is applied to vibrated granular discs, providing evidence of the Gardner phase \cite{64}. In the Gardner phase, one expects a logarithmic growth of the MSD with lag time, which is verified in an experiment of glassy colloidal suspensions \cite{65}. The experimental data of shear modulus and MSD in a hard-sphere colloidal glass are consistent with the scalings $\mu_{ZFC} \sim 1/\Delta_{EA} \sim p^e$ \cite{66}. The critical scalings of weak forces and small interparticle gaps have also been verified by precise experimental measurements of jammed photo-elastic disks \cite{67}. The evidence of a Gardner-like transition is reported in a two-dimensional bidisperse granular crystal \cite{68}, suggesting that the Gardner physics could be observed with minimum disorder \cite{69}. Examining protocol-dependent shear moduli and complex aging dynamics could provide future directions for the experimental exploration of Gardner physics.

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