From Subaging to Hyperaging in Structural Glasses

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We demonstrate nonequilibrium scaling laws for the aging and equilibration dynamics in glass formers that emerge from combining a relaxation equation for the static structure with the equilibrium scaling laws of glassy dynamics. Different scaling regimes are predicted for the evolution of the structural relaxation time τ with age (waiting time t_w), depending on the depth of the quench from the liquid into the glass: "simple" aging ($\tau \sim t_w$) applies for quenches close to the critical point of mode-coupling theory (MCT) and implies "subaging" ($\tau \approx t_w^\delta$ with $\delta < 1$) as a broad equilibration crossover for quenches to nearly arrested equilibrium states; "hyperaging" (or superaging, $\tau \sim t_w^{\delta'}$ with $\delta' > 1$) emerges for quenches deep into the glass. The latter is cut off by non-mean-field fluctuations that we account for within a recent extension of MCT, the stochastic β-relaxation theory (SBR). We exemplify the scaling laws with a schematic model that quantitatively fits simulation data.

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The response of a viscous fluid to a sudden change in control parameters reveals a rich phenomenology as the system adapts to this change. If the timescale of structural relaxation in the fluid τ is large, a slow evolution of both static and dynamic properties of the fluid with system age (i.e., the waiting time t_w after the quench) is observed. For kinetically arrested states such as glasses, this aging dynamics implies that the properties of the material depend on the protocol of its fabrication, a clear nonequilibrium signature [1–3]. The understanding of the nontrivial timescales in aging is fundamental for theoretical physics and materials science alike [4–6].

This concerns, in particular, empirical scaling relations that have been observed in experiment and simulation of systems that are widely different on the microscopic scale, ranging from molecular and polymeric glasses [7–13], colloidal systems [14–17], metallic alloys [18–20], and laponite suspensions [21,22] to spin glasses [23–25]: simple aging ($\tau \sim t_w$) and subaging ($\tau \sim t_w^\delta$ with $\delta < 1$) are commonly found; hyperaging ($\tau \sim t_w^{\delta'}$, with $\delta' > 1$) is present as an intermediate law with a nonuniversal exponent [12,14] and was explicitly reported, for example, in laponite suspensions ($\delta' \approx 1.8$) [26] and colloidal gels ($\delta' \approx 1.37$) [27]. The physical mechanisms behind these scaling laws, and, in particular, their relation to the microscopic details of the fluid and/or the protocol of the control-parameter quench, so far remained unresolved.

Here we establish *nonequilibrium* scaling laws of aging that emerge from the *equilibrium* scaling laws for structural relaxation near the critical point of mode-coupling theory (MCT). They are power laws with nonequilibrium exponents that are, via MCT, directly linked to the equilibrium structure of a glass-forming material and are thus generic, although not universal (since the exponents depend on microscopic details). Our predictions arise from combining two recent theoretical approaches to describe glass-forming fluids: The nonequilibrium self-consistent generalized Langevin equation theory (NE-SCGLE) suggests a starting point linking the evolution of static properties to the relaxation dynamics of the system. The stochastic β-relaxation theory (SBR) provides scaling laws for this relaxation dynamics that also include the effect of non-mean-field fluctuations beyond MCT. We elucidate the predicted aging regimes through quantitative comparison to simulation results for density-quenched hard-sphere-like systems [28,29].

MCT is a microscopic theory [30,31] that successfully describes the liquid-state dynamics close to the glass transition. Originally restricted to the equilibrium ensemble, recent extensions allow one to treat nonlinear response to various external fields [32–36]. Its application to aging dynamics was proposed 20 years ago by Latz [37,38], but the complexity of that theory has so far only allowed to

obtain some results linked to the seminal work by Cugliandolo *et al.* [4,39] on the p-spin model [40]. The complexity stems from the fact that, in absence of the equilibrium fluctuation-dissipation theorem, correlation and response functions are not straightforwardly connected and are described by coupled integral equations that are not readily evaluated.

To cut this Gordian knot, the NE-SCGLE [41–43] invokes an assumption of "local stationarity" for the relaxation process, reducing the complexity of the full problem considerably. Essentially, it partially decouples the evolution of the correlation functions from that of the underlying static response functions. The resulting theory tests favorably against both simulation [28,29,44] and experimental data [45–47].

NE-SCGLE in fact refers to two separate ingredients: an evolution equation for the static observables and an underlying kinetic theory for the mobility of rearrangements, the SCGLE [48]. The latter is, for the present purposes, structurally identical to MCT. In particular, it provides the same robust asymptotic scaling laws for the equilibrium structural relaxation [49]. We will use those well-established scaling laws to describe the asymptotic waiting-time dependence after a quench.

The nonequilibrium extension of the SCGLE usually references Onsager's laws of linear irreversible thermodynamics and the corresponding stochastic theory of thermal fluctuations [50,51]. Under certain assumptions, it leads to an innocuous looking relaxation equation for the waiting-time evolution of the nonequilibrium static structure factor $S(k;t_w)$. We elaborate in the Supplemental Material [52] that this equation can also be rationalized within the integration through transients (ITT) framework used to derive nonequilibrium extensions of MCT [32]. In brief, the nonequilibrium distribution function $p(t)$ obeys $\partial_t p(t) = \Omega(t)p(t)$ with some linear differential operator $\Omega(t)$, and a formal solution is $p(t) - p(t_w) = \int_{t_w}^{t} dt' \exp_+[\int_{t'}^{t} \Omega(\tau)\, d\tau]\Omega(t')p(t_w)$. For a sudden quench, $\Omega(t) = \Omega_i$ for $t < 0$ and $\Omega(t) = \Omega_f$ for $t > 0$; using $\Omega(t')p(t_w) = \partial_{t_w} p(t_w)$ for $t' \geq t_w > 0$ avoids the need to formulate the effect of the quench in the time-evolution operator explicitly. Integrating the formal solution over density-pair fluctuations to obtain the static structure factor, and projecting (in the spirit of MCT) the exponential propagator onto density-pair modes as the relevant variables, we obtain after neglecting memory effects on this level $S(k;t) - S(k;t_w) \approx \int_{t_w}^{t} dt' C_4(k;t,t')\partial_{t_w} S(k;t_w)$ with a four-point density correlation function $C_4(k;t,t')$. Thus, for $t \to \infty$,

$$\frac{\partial S(k;t_w)}{\partial t_w} = -\mu(k;t_w)(S(k;t_w) - S_f(k)), \tag{1}$$

where $\mu(k;t_w)$ is a mobility factor that is slaved to the structural-relaxation dynamics [7,56]. The initial state before the quench is $S(k;0) = S_i(k)$, and $S_f(k)$ characterizes the quenched-to-final state. Equation (1) essentially is a formalized version of the empirical Tool model of physical aging [57].

Equation (1) already predicts universal scaling laws for the aging dynamics to be encoded in the equilibrium dynamics: since the glass transition is a dynamical phenomenon, in its vicinity the static structure functions remain regular, and we can linearize $S(k;t_w)$ for small control-parameter distances $\varepsilon(t_w)$ to the transition. The evolution is thus asymptotically governed by the evolution of the distance parameter along the relevant direction in k space (MCT's critical eigenvector [30,58]),

$$\partial_{t_w}\varepsilon(t_w) = -\mu[\varepsilon(t_w)](\varepsilon(t_w) - \varepsilon_f). \tag{2}$$

Now enter the scaling laws for $\mu(\varepsilon)$: close to the critical point of MCT, $\mu(\varepsilon) \sim 1/\tau(\varepsilon) \sim (-\varepsilon)^\gamma$ for liquid states ($\varepsilon < 0$), and $\mu(\varepsilon) = 0$ in the ideal glass state ($\varepsilon \geq 0$). The exponent γ is related to the equilibrium structure of the system at its glass transition through the MCT exponent parameter λ [30,49,52]. Since μ approaches zero, Eq. (2) has nonequilibrium stationary solutions where the relaxation toward equilibrium gets "stuck."

We immediately get two important scaling laws from Eq. (2).

(i) For quenches close to the glass-transition point ($|\varepsilon_f| \ll |\varepsilon_i|$), there exists a growing window in t_w, where $\partial_{t_w}\varepsilon \sim |\varepsilon|^{\gamma+1}$, which results in $|\varepsilon| \sim t_w^{-1/\gamma}$ and, thus, simple or *full aging*, $\tau \sim t_w$ as $t_w \to \infty$.

(ii) For a deep quench into the ideal glass, $\varepsilon_f \gg |\varepsilon(t_w)|$ holds in the limit of $t_w \to \infty$, because the relaxation gets stuck around values close to zero. Then, $\partial_{t_w}\varepsilon \sim |\varepsilon|^\gamma$, resulting in the asymptotic law $\tau \sim t_w^{\gamma/(\gamma-1)}$. Since $\gamma > 1$, the exponent $\delta' = \gamma/(\gamma-1)$ is also larger than unity, and we find *hyperaging* or superaging, $\tau \sim t_w^{\delta'}$ for $t_w \to \infty$ with a nonuniversal exponent δ' that depends on the microscopic structure of the system.

These scaling laws describe the idealized indefinite aging of a system that is quenched to a state with infinite relaxation time. In reality, the ultimate MCT-like divergence of the relaxation time is not observed. We attribute this to long-wavelength fluctuations that cause deviations from the mean-field-like scenario [59–61] and render the scaling laws transient rather than truly infinite-t_w asymptotes, as we shall discuss below.

For quenches to liquid states close to the glass transition, $\varepsilon_f < 0$, the mobility always remains positive, and the corresponding long-time behavior is then (iii) $\tau \sim$ const for $t_w \to \infty$. For the typical slow evolution of the structural-relaxation time, this implies a broad crossover where τ grows sublinearly with t_w, and hence *subaging* transiently appears during equilibration. Although not a rigorous asymptote, an empirical power law, $\tau \approx t_w^\delta$ with $\delta < 1$, typically fits well in this regime [40].

To elucidate the emergence of the three regimes—simple, sub-, and hyperaging—we devise a schematic model of aging. Qualitatively, the mobility is the inverse of an integrated friction memory kernel; in the spirit of MCT schematic models, we assume that the slow dynamics of all such microscopic correlation functions is governed by a single-mode (density) correlation function $\phi(t;t_w)$,

$$\mu(t_w) = 1/\int_0^\infty dt\,\phi(t;t_w). \tag{3a}$$

The latter obeys a Mori-Zwanzig-type integral equation,

$$\partial_t\phi(t;t_w) + \phi(t;t_w) + \int_0^t m(t-t';t_w)\partial_{t'}\phi(t';t_w)dt' = 0. \tag{3b}$$

In Eq. (3b), we anticipate that t_w only enters parametrically in determining the coupling coefficients of the memory kernel $m(t;t_w)$. This encodes the assumption of local stationarity and is in the spirit of the ITT framework [33] that relates nonequilibrium transport coefficients to such "transient" correlation functions.

We complete the schematic model by the closure

$$m(t;t_w) = v_1(t_w)\phi(t;t_w) + v_2(t_w)\phi(t;t_w)^2, \tag{3c}$$

with two coupling parameters v_1 and v_2 that describe the current t_w-dependent state of the system. For fixed t_w, the model specified by Eqs. (3b) and (3c) is the widely studied schematic F_{12} model of MCT. It has a line of glass transitions (v_1^c, v_2^c) where $\varepsilon = 0$.

Equations (3) define our schematic model. Together with the (mean-field) assumption $\tau(t_w) \propto 1/\mu(t_w)$, and $v_1 = v_1^c$, $v_2(t_w) = v_2^c(1+\varepsilon(t_w))$ to define the distance to the glass-transition point, it allows one to fit available computer-simulation data for $\tau(t_w)$ after mapping $\varepsilon_i = \varepsilon(0)$ and ε_f to the simulation's control parameters.

Results for $\tau(t_w)$ from the schematic model for quenches to various final states close to the MCT transition give a consistent description of computer-simulation data for density-quenched quasihard spheres (Fig. 1). For the fit, we have allowed to adjust a global timescale and the proportionality factor between μ and $1/\tau$. Instead of fitting the exponents to the data, we have fixed the exponent parameter $\lambda = 0.735$ to match the MCT prediction for hard-sphere-like systems [52] (by adjusting v_1^c and v_2^c). This predicts the exponent $\gamma = 1/2a + 1/2b$ with $\Gamma(1-a)^2/\Gamma(1-2a) = \lambda = \Gamma(1+b)^2/\Gamma(1+2b)$, and thus the exponent $\delta' \approx 1.684$.

The schematic model demonstrates the three aging regimes of the theory: empirical subaging is found as an equilibration crossover for quenches to final states in the liquid, $\varepsilon_f < 0$, while hyperaging emerges as the asymptote for quenches to the glass, $\varepsilon_f > 0$. A growing intermediate-t_w window that extends to $t_w \to \infty$ at the critical point of MCT, $\varepsilon_f = 0$, displays simple aging.

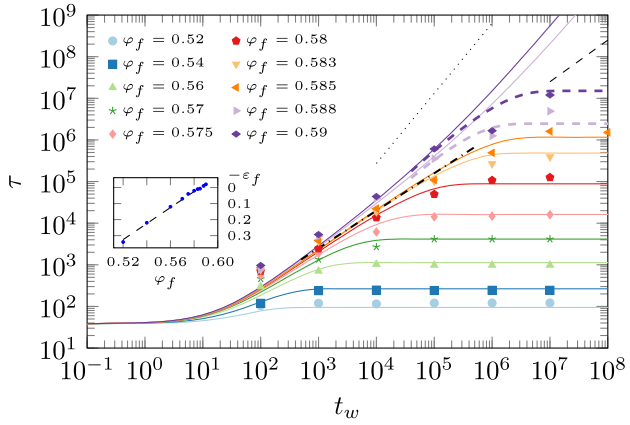

FIG. 1. Structural-relaxation time τ as a function of waiting time t_w after an instantaneous quench. Solid lines: schematic model, quenches from $\varepsilon_i = -0.5$ to ε_f. Inset: values in the Supplemental Material [52]. A dashed line indicates simple aging $\tau \sim t_w$, a dotted line hyperaging $\tau \sim t_w^{\delta'}$ with $\delta' = 1.684$, and a dash-dotted line subaging $\tau \approx t_w^{\delta}$ with $\delta = 0.9$. Thick dashed lines: SBR for $\varepsilon_f = 0.01$ and 0.02. Symbols: simulation results for quasihard spheres from Ref. [29], quenched to various final packing fractions φ_f, translated to schematic-model units ($\tau \mapsto 2\tau$, $t_w \mapsto 100 t_w$).

The evolution of τ after the quench relates to the well-known problem of determining a diverging relaxation time at fixed waiting time t_w (corresponding to a typical experiment duration or probing timescale): approaching the transition, the power-law divergence of τ as a function of quenched to state ε_f that is predicted by the idealized theory, is cut off at any finite t_w and replaced by a crossover to a slower growth (Fig. 2). In our theory, we obtain $\tau \sim |\varepsilon|^{\delta'}$, with a prefactor that diverges with increasing t_w (dash-dotted lines in Fig. 2).

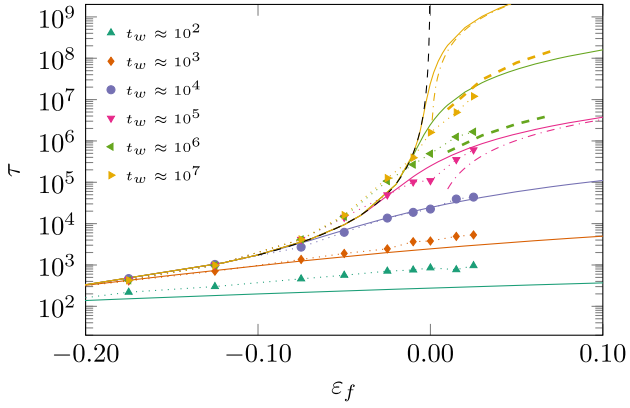

FIG. 2. Structural-relaxation time τ at various fixed waiting times t_w as a function of the final point of the quench ε_f (solid lines and symbols: theory and simulation as in Fig. 1; thick dashed lines from SBR). Thin dashed line, equilibrium divergence at the ideal glass transition point ($\varepsilon_f = 0$), $\tau_{\rm eq} \sim |\varepsilon_f|^{-\gamma}$ ($\gamma = 2.46214$ corresponding to $\lambda = 0.735$); dash-dotted lines, nonequilibrium asymptotes $\tau \sim B(t_w)\varepsilon_f^{\delta'}$.

Deviations from the ideal theory are noted in the simulation data for quenches to the highest final densities and at large t_w. We attribute this to the avoidance of the ideal MCT transition, which also causes the hyperaging regime to be interrupted.

To understand this, we turn to the SBR [59,60], a recent extension of MCT that includes fluctuations in the local glassiness, viewing $\sigma \sim \varepsilon$ as a dynamical fluctuating order parameter. SBR predicts scaling laws that replace the divergent power law with a crossover between a power law and exponential growth. Specifically [62], different laws arise for the structural-relaxation time and for the mobility,

$$\tau \sim \left[\int_{-\infty}^{0} \frac{ds}{\sqrt{2\pi}\Delta\sigma} e^{-\frac{(s-\sigma)^2}{2\Delta\sigma^2}} |s|^{b\gamma}\right]^{-1/b}, \tag{4a}$$

$$\mu \sim \int_{-\infty}^{0} \frac{ds}{\sqrt{2\pi}\Delta\sigma} e^{-\frac{(s-\sigma)^2}{2\Delta\sigma^2}} |s|^{\gamma}, \tag{4b}$$

where we identified $\sigma = \varepsilon$. Here, $\Delta\sigma$ is a material parameter that quantifies the strength of long-wavelength order-parameter fluctuations (we set $\Delta\sigma = 0.1$). Using Eq. (4) to evaluate μ in Eq. (2) and to calculate τ, we obtain an improved asymptotic description of the τ versus t_w curves (colored dashed lines in Fig. 1) that account for the crossover from hyperaging to a constant τ as the system finally equilibrates even in the ideal MCT glass.

A clear hyperaging signature still survives as a transient. In the simulation data, this is best seen as a nonmonotonic variation of the ratio τ/t_w as a function of t_w that is present for quenches to $\varphi_f > \varphi_c$ (Fig. 3) and that fits well the corresponding SBR prediction (solid lines in Fig. 3). Experimental data on colloidal suspensions [14] for deeper quenches show the nonmonotonic variation of t/τ_w even more clearly (see Supplemental Material [52]).

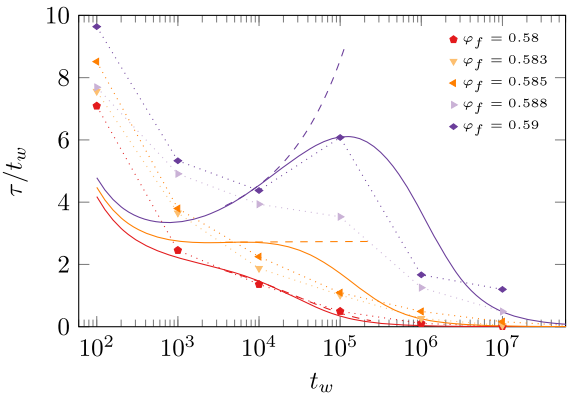

FIG. 3. Ratio τ/t_w as a function of waiting time t_w. Simulation data of Ref. [29] (filled symbols, data divided by 50; $\varphi_c \approx 0.585$) and exemplary SBR results for $\varepsilon_f = -0.1$, 0, and 0.1 (solid lines, bottom to top). Dashed lines indicate the corresponding asymptotes of the ideal glass MCT.

It is worth noting that the qualitative change observed in Figs. 1 and 3—true aging replaced by eventual equilibration—implies a merely quantitative change for a measurement performed at fixed system age: SBR predictions for τ at fixed t_w (dashed lines in Fig. 2) are only shifted compared to the ideal MCT curves. Experiments at fixed system age can never address the question of whether τ truly diverges or not.

To summarize, we obtained scaling laws for the evolution of the structural relaxation time τ as a function of system age t_w after the quench of a glass-forming fluid to states close to the ideal glass-transition point of MCT. The scaling laws delineate regimes of simple and transient hyper- and subaging.

The results link the hyperaging exponent δ' to the exponent characterizing the equilibrium relaxation time. Hence, they link a nonequilibrium dynamical exponent of the system to a nontrivial equilibrium exponent and through this to the system's equilibrium static structure.

We conclude with some remarks. (i) Our analysis based on Eq. (1) takes any theory of glassy dynamics as input. Like the ITT framework, it can, in principle, be combined with approaches different from MCT or SBR, although the derivation of the scaling laws from Eq. (2) is more rigorous within MCT where glass transitions are A_ℓ bifurcations with a single critical eigenvector in k space.

(ii) The predicted hyperaging exponent δ' is *not* universal; it depends on the details of the microscopic structure. It can be determined from the exponent parameter $\lambda \in [1/2, 1]$ through a scaling analysis of the equilibrium dynamics, and within MCT it is calculated via a functional of the equilibrium static structure factor $S(q)$ (see Supplemental Material [52] for details). In practice, generic values of λ (and δ') emerge for broad classes of glass formers: $\lambda \approx 0.735$ fixes $\delta' \approx 1.68$ as typical for hard-sphere-like glasses. On the boundary between repulsive and attractive glasses in systems with strong short-ranged attractions, higher-order glass-transition singularities [63] are predicted to suppress hyperaging (as there, $\gamma \to \infty$ and hence $\delta' \to 1$); the value $\delta' \approx 1.37$ found in Ref. [27] corresponds to $\lambda \approx 0.88$, which is not unreasonable for attractive glasses predicted by MCT. Systems with nearly nonstretched structural-relaxation dynamics, on the other hand, should approach the upper limit of $\delta' \approx 2.3$.

(iii) The connection between equilibrium and nonequilibrium dynamics rests on the β-scaling law, a very robust feature of MCT that emerges also in a more general field-theoretical framework [61] or in generalized mode-coupling theory [64]. It will be interesting to see how full microscopic calculations beyond the schematic model will reflect on the predicted aging behavior.

(iv) Interrupted hyperaging versus subaging emerges as a clear separation between ideal glasslike dynamics and the dynamics that arises from the avoidance of the ideal glass transition. Hyperaging is also predicted to be more

pronounced as one quenches deeper into the glass. This leads us to speculate that models with a nonavoided MCT-like glass transition might show clear long-time hyperaging asymptotes. High-dimensional systems of hard spheres, approaching the expected mean-field-like behavior in $d = \infty$ [65,66], could be suitable candidates. Numerical solutions of spin glasses with MCT transitions, e.g., the spherical p-spin model, so far favor sub- and normal aging [4,40,67], but the analytical determination of the scaling laws is still an open issue in these models [40,67]. Hyperaging in a trapped phase has been discussed very recently in the context of decision-making models that incorporate reinforcement by memory effects [68]. Our Eq. (2) predicts weak ergodicity breaking and aging that gets stuck at the MCT-critical point; it will be interesting to explore the connection to the strong ergodicity breaking discussed in spin glasses [69] and the loss of ultrametricity connected with the hyperaging asymptote in suitably enhanced models.

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