

Time glass: A fractional calculus approachR. C. Verstraten¹, R. F. Ozela^{1,2} and C. Morais Smith¹¹*Institute for Theoretical Physics, Utrecht University, Princetonplein 5, 3584CC Utrecht, The Netherlands*²*Faculdade de Física, Universidade Federal do Pará, 66075-110 Belém, PA, Brazil*

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Out-of-equilibrium states in glasses and crystals have been a major topic of research in condensed-matter physics for many years, and the idea of time crystals has triggered a flurry of new research. Here, we provide a description for the recently conjectured time glasses using fractional calculus methods. An exactly solvable effective theory is introduced with a continuous parameter describing the transition from liquid through normal glass and time glass into the Gardner phase. The phenomenological description with a fractional Langevin equation is connected to a microscopic model of a particle in a sub-Ohmic bath in the framework of a generalized Caldeira-Leggett model.

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Introduction. The concept of time crystals, i.e., materials with an emergent periodicity in time [1,2] has attracted much attention recently [3]. After no-go theorems were proven for several systems [4,5], it was understood that time crystals may actually occur in open [6,7], driven [8], and long-range interaction [9] systems, and many experimental and theoretical activities followed this novel idea. By now, time crystals have been conceived and observed [10–14]. Recently, a novel system was conjectured, a time glass, which would have periodic intermediate glass states [15]. It was claimed that they should appear in static many-body localized systems exhibiting incommensurate local frequencies and no long-range spatiotemporal order [3].

The microscopic states of glasses have been puzzling researchers for many years [16–21]. Microscopically, glasses look like liquids because their molecules do not show any kind of structural order. These amorphous materials exhibit completely different phase transitions in comparison to ordered solids, and often they do not even have a thermodynamic ground state. Hence, equilibrium physics cannot be used for their description [22]. A common understanding is that they correspond to the occupation of a set of metastable states in the free-energy landscape.

One promising approach to describe the glass transition is the random first-order transition theory [23]. It describes a hard-sphere model in which the spheres can get caged by their neighbors, thus, restricting their movement. This leads to a mean-square displacement (MSD) that corresponds to a free particle at small timescales, until it hits the cage size (see Fig. 1), where the MSD saturates, indicating a glassy state [24,25] (for a discussion on glass states, see the Supplemental Material (SM) [25–31]). In the free-energy landscape, this is the moment at which the particle has explored the entire basin. This theory, however, can only be solved exactly in an infinite dimension with a questionable connection to its finite-dimensional counterpart [32].

More recently, a richer phase diagram was proposed for glasses, including the so-called Gardner phase [33,34].

Although the Gardner phase was first discovered in the description of spin glasses as a solution that breaks one replica symmetry [35], it was later understood to occur in many materials [36]. This marginal glass phase has a free energy in which basins transform into metabasins [37] and is known to have a fractal structure [38] (see Fig. 1). In the Gardner phase, there is a hierarchy of cages inside cages [39], reminiscent of the fractal structure in the energy landscape. Therefore, as time goes by, the system explores larger cages, thus, triggering an infinite staircaselike behavior of the MSD.

Here, we propose a unified picture where liquid, glass, time glass, and the Gardner phase can be understood as a (sub-)diffusive Brownian motion described by a fractional Langevin equation with white noise. Previously, the fractional Langevin equation had been studied for colored noise [40–51], but the features described here occur exclusively for the case of white noise. At very low temperatures, the equipartition theorem breaks down, and a white-noise fractional Langevin equation arises as the semiclassical description of a particle coupled to a two-level systems (TLS) bath in the subohmic regime. Our unified procedure has a single varying generic s -derivative friction (with s integer or fractional) to describe the different states of matter. Moreover, we show that for $0 < s \lesssim 0.1$, a *time glass* emerges with a periodicity of $\pi(M/\eta)^{1/(2-s)}$, where M is the mass of the “Brownian” particle and η is a type of viscoelasticity. Within our mean-field description, the time glass shows an emergent frequency and periodically passes through many metastable states before it gets to a final frozen glassy state. Therefore, our Letter sets a mathematical framework for the description and realization of a time glass, thus, furthering our understanding of “time materials.”

Fractional calculus. Fractional differential equations have been used in physics, engineering, material science, control systems, protein folding [40], and more [52], but there are still many new opportunities to be explored [41]. Different definitions were proposed by Riemann-Liouville, Caputo, Weyl, etc. (see the SM [26,53–59] for a short historical overview).

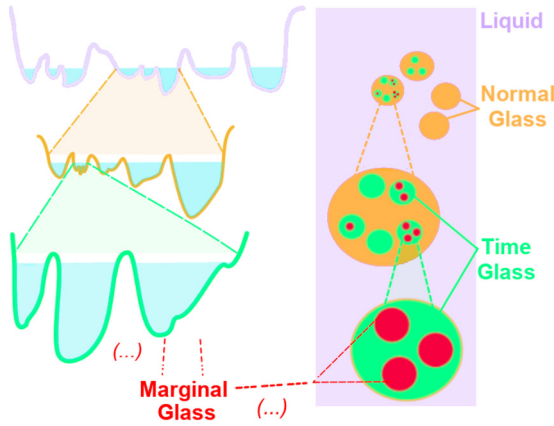


FIG. 1. Left: Sketch of a free-energy landscape with fractal-like metastable minima. Right: A hierarchy of cages inside cages. The energy landscape (purple) has four local minima, corresponding to the total number of large orange cages. Zooming in on one of these minima, we see five smaller local minima (green cages) inside this orange cage. Moreover, inside one green cage there are three red cages. Depending on the number of nested cages we find four differing phases: Liquid (no cage), normal glass (one cage), time glass (finite number of cages), and marginal glass (infinite number of cages).

From a mathematical perspective, there is still a discussion on which of the various fractional derivative definitions [60] should be used for each kind of problem. Concerning the Caputo definition, the idea is to rewrite a repeated integral into a generalizable form. As factorials often appear in conventional integrals, one uses the Γ function as their noninteger generalization. The Caputo derivative is then given by taking an integer derivative before doing a fractional integral,

$$\mathbf{D}_t^\alpha f(t) = \frac{1}{\Gamma(n-\alpha)} \int_0^t (t-\tau)^{n-\alpha-1} f^{(n)}(\tau) d\tau, \quad (1)$$

where n is an integer such that $n-1 \leq \alpha < n$. Since the Caputo definition is nonlocal, we have chosen the left-handed definition for the boundary of the integral to retain causality, once we apply this time derivative to our system. One benefit of the Caputo definition compared to other definitions is that we can keep integer-order boundary conditions; however, continuity in the order is lost on the integers. When this noninteger derivative is applied to an exponential, we find the Mittag-Leffler function, defined by

$$E_{\alpha,\beta}(t) = \sum_{k=0}^{\infty} \frac{t^k}{\Gamma(\alpha k + \beta)}, \quad (2)$$

which is a generalized exponential that appears regularly in solutions of fractional differential equations. For different parameters, this function can show many features related to exponentials, such as damped oscillations and exponential-like growth.

Fractional Langevin equation. The fractional Langevin equation was recently used to describe a system exhibiting Lévy flights [42]. Lévy flights are often used for modeling the spreading of viruses as they include a description of the long (and fast) journeys that people make by plane as well as

a more local random motion. Brownian motion only has one typical time and length scale associated with it, whereas Lévy flights have many different time and length scales [40,61]. The equipartition theorem implies a relation between the fluctuation and the dissipation terms [62]; hence, previous references used either colored noise with the fractional Langevin equation [40–46] or a white noise associated with a fractional kinetic term and normal friction [63] which, after fractional integration, is equivalent to the fractional Langevin equation with colored noise. However, none of these models exhibit the plateaus in the MSD characterizing a glassy behavior, which we will describe below. The features that we will discuss here are inherent to a fractional Langevin equation with white noise and occur only at low temperatures when the equipartition theorem breaks down. Later, in the *Microscopic model* section, we present a microscopic description corresponding to a physical realization of our phenomenological model.

Let us start by considering the *fractional Langevin equation*,

$$M \frac{d^2 x(t)}{dt^2} + \eta \mathbf{D}_t^s x(t) = f(t), \quad (3)$$

with $f(t)$ as a white-noise force with average $\langle f(t) \rangle = 0$ and correlation $\langle f(t)f(t') \rangle = K\delta(t-t')$, where $K = 2 \sin(\frac{\pi s}{2}) \eta t_s^{1-s} k_B T$, and $t_s = (M/\eta)^{1/(2-s)}$ is the timescale of the system. Introducing these fractional derivatives as friction, scaling with the s th order derivative, yields a model for both subdiffusion ($s < 1$) and superdiffusion ($s > 1$) in addition to the usual Langevin equation, which is retrieved when $s = 1$. The motivation for this change in friction compared to the Langevin equation is first to introduce a general nonlocal operator that knows about the history of a system and allows one to study non-Markovian processes. The choice to take fractional derivatives can then be illustrated with a thought experiment: Suppose a particle is moving at a constant speed for a certain time. Then, ordinary friction is constant in time, whereas friction of this fractional form scales as t^{1-s} . This means that for $s > 1$ the friction will fall off quickly, allowing for Lévy flights, whereas for $s < 1$ the friction will increase in time, thus, reducing the probability for large jumps [64].

The strength of this theory is that it is exactly solvable, which makes possible the calculation of statistical properties, such as the MSD. For $0 \leq s < 1$, the MSD is given by (see the SM [26,65,66] for details)

$$\langle x(t)^2 \rangle = \frac{K}{M^2} \int_0^t \left[\tau E_{2-s,2} \left(-\frac{\eta}{M} \tau^{2-s} \right) \right]^2 d\tau + \left[v_0 L_s \frac{t}{t_s} E_{2-s,2} \left(-\frac{\eta}{M} t^{2-s} \right) \right]^2, \quad (4)$$

where $v_0 = \langle x'(0) \rangle t_s / L_s$ is dimensionless. The short-time expansion for $t \ll t_s$ yields a ballistic behavior,

$$\langle x(t)^2 \rangle = (v_0 L_s)^2 \left(\frac{t}{t_s} \right)^2, \quad (5)$$

whereas the long-time expansion is logarithmic for $s = 0.5$, and otherwise given by

$$\langle x(t)^2 \rangle = \Delta_s^2 + \frac{K t^{2s-1}}{(2s-1)\eta^2 \Gamma(s)^2}, \quad (6)$$

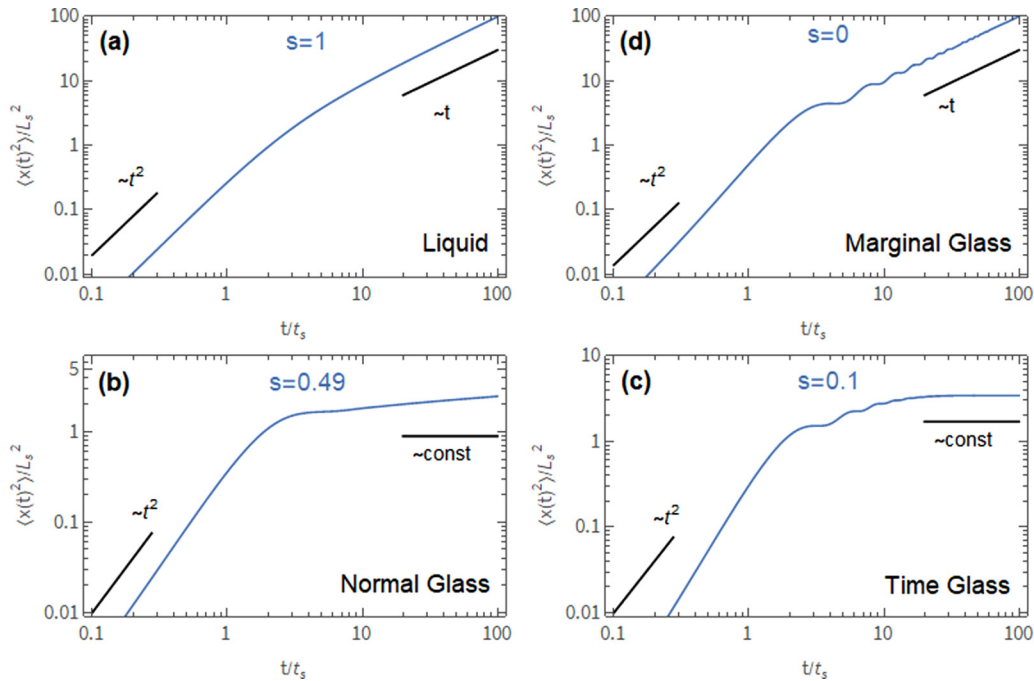


FIG. 2. The exact MSD for different values of s , regularized by the typical time and length scales t_s and L_s . The analytical asymptotes for $t \rightarrow 0$ and $t \rightarrow \infty$ are drawn next to the plots. The delocalized states are shown at the top and the localized states at the bottom, whereas the labeling order follows the decreasing value of s . (a) A liquid, described by regular Brownian motion. (b) A normal glass, exhibiting ballistic motion at short times and localization at longer times. (c) A time glass, displaying ballistic motion at short times, but also an intermediate regime with a set of small plateaus before the long-time plateau sets in. (d) A marginal glass, characterized by a driven particle in a quadratic potential. Here, the friction term vanishes and an infinite collection of plateaus appear. The overall slope is however finite, reminiscent of a liquid behavior.

where, for $s < 1/2$, the exponent of the second term in the MSD becomes negative and, therefore, the MSD converges to the typical final cage size Δ_s^2 for $t \rightarrow \infty$.

In Fig. 2, the MSD has been plotted for several values of s from one to zero. We introduced the typical length scale of the system, which can be found by dimensional analysis to be $L_s = \sqrt{Kt_s^3/M^2}$. The MSD shows ballistic short-time behavior in all cases. For $s = 1$, we retrieve the conventional Langevin equation, which describes Brownian motion. The MSD also shows a crossover from a ballistic ($\sim t^2$) to a linear dependence in time, characteristic of a liquid [Fig. 2(a)]. For $s \lesssim 0.5$, instead, the MSD saturates at long times, thus, describing a glass [Fig. 2(b)]. We find that a particularly interesting regime is provided by small values of s in the interval $0 < s \lesssim 0.1$. In this case, a sequence of small metastable plateaus characterizes a finite-depth fractal glass phase before the conventional glass regime is reached at longer times [Fig. 2(c)]. For $s = 0$, the “marginal glass” phase, proposed by Gardner, is realized with an infinite number of metastable plateaus and finite average slope ($\sim t$), typical of liquids. This is an asymptotic phase in which the fractal glass acquires infinite depth [Fig. 2(d)].

Time glass. Recently conjectured [15], *time glass* is a phase in which the MSD periodically repeats a glasslike plateau in an overall glassy phase (see the SM [26] for a comparison between definitions of time glass). Now, we concentrate on the region $0 < s \lesssim 0.1$, which describes a finite-depth fractal glass, reminiscent of the Gardner phase (see Fig. 1). The evolution of the MSD upon varying s is depicted in Fig. 3

and discloses many interesting traits: (i) At short times ($0 < t < \pi t_s$), there exists a universal regime in which all curves collapse into a single one; (ii) the small plateaus regime sets in afterwards, but the overall slope of the intermediate-time behavior *increases* as s is reduced, thus, showing a gradual transition from an overall glass to liquid phase; (iii) at sufficiently long times, there is saturation for all $s \neq 0$, but this freezing occurs on increasingly longer timescales as s is reduced; (iv) these plateaus appear at fixed intervals in time as

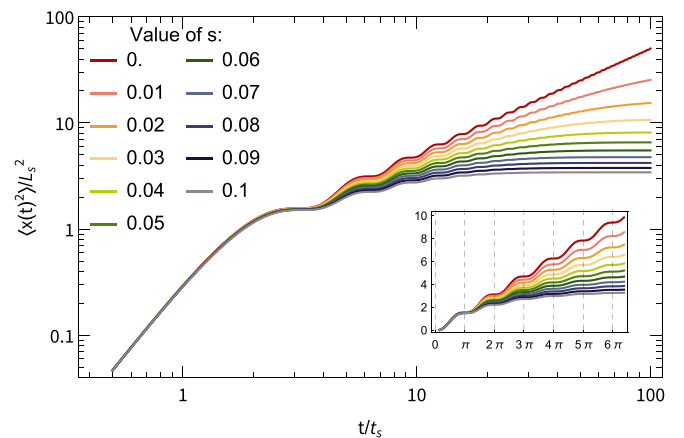


FIG. 3. The MSD for several small values of s , regularized by the typical time and length scales t_s and L_s . The inset shows a linear scale plot to highlight the periodicity in the anomalous glass phase, characterizing a time glass.

promptly visualized in a linear scale plot (inset of Fig. 3). Put together, all these features closely resemble (see the SM [26]) the marginal glass phase that has recently been observed in colloidal glass experiments [67] and the emergent frequency that we obtain indicates that we are describing a *time glass* phase. These results are further corroborated by calculations of the position and velocity autocorrelation functions, which show the same periodicity (see the SM [26]).

To find the coefficient of the timescale such that we have the period of the system, we concentrate on the case $s = 0$ and put $v_0 = 0$ for simplicity. As this case is just a driven harmonic oscillator, we get

$$\begin{aligned} \langle x(t)^2 \rangle &= \frac{K}{M^2} \int_0^t \underbrace{\left[\tau E_{2,2} \left(-\frac{\eta}{M} \tau^2 \right) \right]^2}_{=\sin^2(\sqrt{\frac{\eta}{M}} \tau)} d\tau \\ &= \frac{K}{2M\eta} \left[t + \frac{1}{2} \sqrt{\frac{M}{\eta}} \sin \left(2\sqrt{\frac{\eta}{M}} t \right) \right], \end{aligned} \quad (7)$$

where the relation between the Mittag-Leffler function and the sine can be seen using their Taylor expansions. This yields a periodicity of $\pi\sqrt{M/\eta}$ in time, thus, providing the general period $P = \pi(M/\eta)^{1/(2-s)} = \pi t_s$ of the system. For nonzero s , this period applies only in a finite time window before freezing.

Microscopic model. Now, we aim at identifying an underlying microscopic model, which is phenomenologically described by the fractional Langevin equation. We will consider an open quantum system, upon which we perform a mean-field approximation to obtain an effective model. Since Hamiltonian dynamics relies on conservation of energy, we have to couple a system undergoing friction to a bath, which exerts that force. Inspired by a generalization of the Caldeira-Leggett model [68–73], we consider a Hamiltonian,

$$H = H_S + H_B + H_{\text{int}}, \quad (8)$$

where

$$H_S = \frac{\hat{p}^2}{2M} + V(x) \quad (9)$$

is a Hamiltonian describing a Brownian particle with mass M , momentum p , coordinate x , and subject to a potential $V(x)$,

$$H_B = \sum_j \frac{\hbar\omega_j}{2} \sigma_{zj} \quad (10)$$

describes a TLS bath (i.e., truncated harmonic oscillators) with natural frequencies ω_j and

$$H_{\text{int}} = -x \sum_j J_j \sigma_{xj} \quad (11)$$

is the interaction between the bath and the Brownian system [68]. The degrees of freedom in the bath are then integrated out to describe quantum dissipation in the system. The spectral function is given by the imaginary part of the Fourier transform of the retarded dynamical susceptibility of the TLS bath, namely,

$$J(\omega) := \text{Im} \mathcal{F}(-i\Theta(t-t')[F(t), F(t')]), \quad (12)$$

and it is crucial for connecting the microscopic parameters of the Hamiltonian with the phenomenological viscoelasticity coefficient η appearing in the Langevin equation. Here, $F(t)$ is the force produced by the particle on the bath. For an Ohmic bath of harmonic oscillators, the spectral function is given by $J(\omega) = \eta\omega$ for $\omega < \Omega$, where Ω denotes a cutoff frequency and is zero otherwise [74]. Such a bath will then be effectively described by the Langevin equation where the friction is proportional to the velocity (first derivative of position) of the system. However, for our choice of sub-Ohmic TLS bath [69,75], the spectral function is given by

$$J(\omega, T) = \eta \sin\left(\frac{\pi s}{2}\right) \omega^s \tanh\left(\frac{\hbar\omega}{2k_B T}\right) \Theta(\Omega - \omega), \quad (13)$$

where $0 < s < 1$ and Ω is a cutoff frequency. The hyperbolic tangent is important for the quantum description where it allows the temperature to select which bath components are more relevant, namely, the ones with frequencies $\omega > 2k_B T/\hbar$. In our semiclassical approach, however, we take the limit $k_B T \ll \hbar\Omega$, which reduces it to the Caldeira-Leggett model, apart from losing equipartition. We can, therefore, rescale the TLS energies by $\omega^{1-s} t_s^{1-s}$ to retain a white-noise correlation. The friction force is then given by

$$F_{fr} = \frac{2}{\pi} \frac{d}{dt} \left\{ \int_0^t \int_0^\infty \frac{J(\omega)}{\omega} \cos[\omega(t-t')] q(t') d\omega dt' \right\}, \quad (14)$$

which, after a reparametrization $\omega \rightarrow \omega/(t-t')$, becomes proportional to a fractional derivative in the limit $k_B T \ll \hbar\Omega$. The proportionality constant is then calculated by a complex contour integral, which results in a finite value for $0 < s < 1$ (see the SM [26] for details of the calculation). This then leads to a friction term given by

$$F_{fr} = \eta \mathbf{D}_t^s q(t). \quad (15)$$

Therefore, a particle interacting with a sub-Ohmic two-level systems bath is well described by the fractional Langevin equation with white noise in the low-temperature limit.

Conclusion. Making use of fractional Caputo derivatives, we have shown that a semiclassical system coupled to a low-temperature sub-Ohmic bath of two-level systems can be described by the fractional Langevin equation with white noise. We have solved this equation analytically and analyzed the anomalous diffusion.

Different behaviors were observed, depending on s , from ordinary Brownian motion for $s = 1$ to glassy behavior for $s \lesssim 0.5$, a time glass for $0 < s \lesssim 0.1$, and a marginal glass for $s = 0$. Our Letter extends the use of fractional derivatives to the realm of subdiffusion by linking the formalism to the description of the Gardner transition. We identified a new regime between the Gardner phase and the usual glass and showed that it is a realization of the long sought time glass. Further extensions of this model using the techniques applied to the fractional Langevin equation with colored noise [47–49] are anticipated.

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