

# Rare Cage Escapes Drive Relaxation in Deeply Supercooled Liquids

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The relaxation dynamics of glass-forming liquids shows at a temperature  $T_c$ , somewhat above the glass-transition temperature  $T_g$ , a crossover, indicating the conjunction of two different dynamical regimes. For temperatures slightly above  $T_c$ , experiments and computer simulations have extensively probed this dynamics on the particle level and identified several universal scaling laws that are often compatible with the predictions of ideal mode coupling theory, while for temperatures below  $T_c$ , the nature of the relaxation dynamics, and hence the reason for the crossover, has so far remained elusive. Here we use large-scale computer simulations to investigate how in the low-temperature regime the relaxation mechanism differs from the one at higher temperatures. Our analysis reveals unexpected scaling laws that allow us to give a surprisingly simple description of the relaxation dynamics at very low  $T$ 's. Space-time correlation functions show that the cage-escape process involves rare but large particle displacements. These displacements give rise to distinctive subdiffusive power laws in the mean squared displacement and the intermediate scattering function, and also rationalize the origin of the universal excess wing found in the dynamic susceptibility. This insight advances our understanding on the dynamics of glass-forming systems at temperatures that are close to the experimental glass transition.

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## I. INTRODUCTION

Impressive advancement in our understanding of glassy systems has been made in the past decade, progress that was triggered by novel theoretical and technical tools like machine learning and advanced simulation techniques, as well as by experimental methods that have allowed us to probe the static and dynamic properties of supercooled liquids and glasses in unprecedented detail [1–8]. These studies have permitted us to reach, on the particle level, a satisfying theoretical understanding of the relaxation dynamics of glassy systems for temperatures that are around or above the critical temperature  $T_c$  of mode coupling theory (MCT) [9,10].

Much less is known, however, about the dynamics below  $T_c$  since at these temperatures the relaxation times are usually by many orders of magnitude larger than the

vibrational timescales, making it very challenging to examine the details of the  $\alpha$  relaxation. (Although network glass formers can be simulated also below  $T_c$  [11], their relaxation dynamics is very different from the one of fragile glass formers [12–14].) Also from the theoretical side, the sub- $T_c$  regime is not well understood, since it is difficult to make analytical predictions that go beyond the predictions made by the simplest version of MCT [15–19], while so far the nonlinear Langevin equation theory calculations make only predictions on the dynamics of a tagged particle [20–22]. Although there is consensus that close to and below  $T_c$  the divergence of the relaxation time as predicted by MCT must be cut off [10,18,23–31], the details of the process governing the  $\alpha$  relaxation, often called “hopping process” [27,32–34], have barely been elucidated since standard simulation techniques make it difficult to access the temperatures significantly below  $T_c$ .

Important advances in simulation techniques—such as the Monte Carlo swap algorithm [3,5,35–37]—have recently opened the door to equilibrate glass systems at ultracold temperatures, permitting us to investigate the static and thermodynamic properties of the glass former [38]. Although such approaches allow us to equilibrate the samples at low temperatures, the number of decades in time over which the dynamics can be probed is not increased,

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resulting that so far very few works have addressed the particle dynamics at low temperatures [3,39–43] and most of these studies considered only relatively modest system sizes, thus neglecting finite size effects that are known to become important for the dynamics at low temperatures [44,45]. In addition, the swap Monte Carlo algorithm has the drawback that it can be applied only to a very small subclass of glass formers, notably systems in which the particles have a substantial polydispersity. This raises the question of whether the results obtained so far for the dynamics are generic, or are affected strongly by the polydispersity of the system [42].

In view of the mentioned drawbacks of previous studies (insufficient decades in dynamics and/or system sizes with less than a few thousand particles [39,43] and that are hence potentially affected by finite size effects [44,45]), the present work specifically addresses the question of determining the relaxation process in a temperature range that allows us to probe how the dynamics changes from an MCT-dominated regime to a sub- $T_c$  dynamics in which the complete establishment of *deeply supercooled conditions* is reached [38]. To this aim, we use a paradigmatic model of generic glass former [28,46–49], with a polydispersity (around 8%) that is weak compared to that typically adopted in swap dynamics [3,39,43], and a system size that is sufficiently large to avoid finite size effects. These large-scale MD simulations are performed over a time span with few similar precedents in the literature (for comparisons, see the recent works Refs. [24,25,38,42,43]). Specifically, our system is a binary Lennard-Jones mixture, a model of supercooled liquid often used in the past to study the relaxation dynamics of simple glass formers [48], with a slight modification of the interaction parameters that suppresses crystallization [50], but does not affect the (above  $T_c$ ) relaxation dynamics; i.e., the critical temperature  $T_c$  remains close to the value of the original model,  $T_c \approx 0.435$  [48]. Note that, in our binary mixture, the majority component is the relevant constituent for the slow dynamics. Simulations were performed at constant density in a wide temperature range, spanning from “hot liquid” conditions to deeply supercooled ones (down to  $T \approx 0.86T_c$ ). In the following we will use reduced units, with  $\sigma$  and  $\epsilon$  of the Lennard-Jones potential as units for length and energy, respectively, while time is expressed in units of  $\sqrt{m\sigma^2/\epsilon}$ , where  $m$  is the mass of a particle. More details on the simulations are given in Sec. IV.

Our findings demonstrate that around and below  $T_c$  glassy dynamics displays novel universal aspects that are directly related to a change of the particle-level transport mechanism occurring around  $T_c$ . Our characterization of this new type of motion permits us to rationalize in a quantitative manner the nature of the so-called slow  $\beta$  relaxation, a dynamical feature widely observed in the low-temperature dynamics of glass-forming liquids and

which gives rise to an excess wing in the dynamic susceptibility [51].

## II. RESULTS

### A. Qualitative change of the particle dynamics around the mode coupling temperature

Figure 1(a) presents  $\langle r^2(t) \rangle$ , the mean squared displacement (MSD) of a particle, as a function of the rescaled time  $t/\tau_F$ , where the Fickian timescale  $\tau_F$  [53] is defined as  $\tau_F = \sigma^2/6D$ , with  $D$  the diffusion constant obtained from the MSD at long times. [The unscaled data for the MSD are shown in Fig. S1a in Supplemental Material (SM) [58].] By construction, this rescaling makes that in the diffusive regime all MSDs fall on a master curve. For  $T$ 's slightly above  $T_c$ , the data follow a master curve also in the time range in which the particles leave the plateau, i.e., at the end of the caging regime. This behavior is compatible with MCT, which predicts for the MSD a  $t$  dependence of the form

$$r_c^2 + a(t/\tau_F)^b + c(t/\tau_F), \quad (1)$$

with a cage size  $r_c$  and exponent  $b$  that are independent of  $T$ ; see Fig. S2 [58] for details on this master curve. Previous studies indicated  $r_c \approx 0.19$  and  $b \approx 0.5$  [46], fully compatible with the present results, see Fig. S3 [58]. Interestingly, one finds that for temperatures below  $T_c$  the MSDs no longer fall onto a master curve, indicating that the escape of the particles from the cage is related to a microscopic mechanism that differs from the one at higher  $T$ 's. The height of the plateau, obtained as indicated in Sec. IV, now depends on  $T$ ; see inset in Fig. 1(a). Since above  $T_c$  the cage size is constant, we conclude that above and below  $T_c$  the  $T$  dependence of  $r_c(T)$  is very different, reflecting the qualitative change of the dynamics even on the timescales of the vibrational motion. (In Fig. S3 we present the  $T$  dependence of  $r_c$  in the full temperature range, demonstrating the marked change close to  $T_c$  [58].) This dependence is linear in  $T$ , at variance with the  $T^{1/2}$  dependence expected for a harmonic system. This result demonstrates that the cage has a significant anharmonic shape [33], in agreement with previous studies on the configurational entropy [54], the predictions from dynamical mean field calculations [21], and with the result that  $T_c$  corresponds to a transition in the dynamic exploration of the potential energy landscape [55–57]. Note that in Fig. 1(a) [or in Fig. 1(b) discussed below] we do not show curves for temperatures that are very close to  $T_c$ , since their presence obscures the fact that there are two different master curves. In Fig. S1 we show the same type of graph, but also including curves at intermediate temperatures [58].

Figure 1(b) shows that also the self-intermediate scattering function  $F_s(q^*, t)$  (SISF) displays the same qualitative scenario emerging from Fig. 1(a) when time is scaled by the

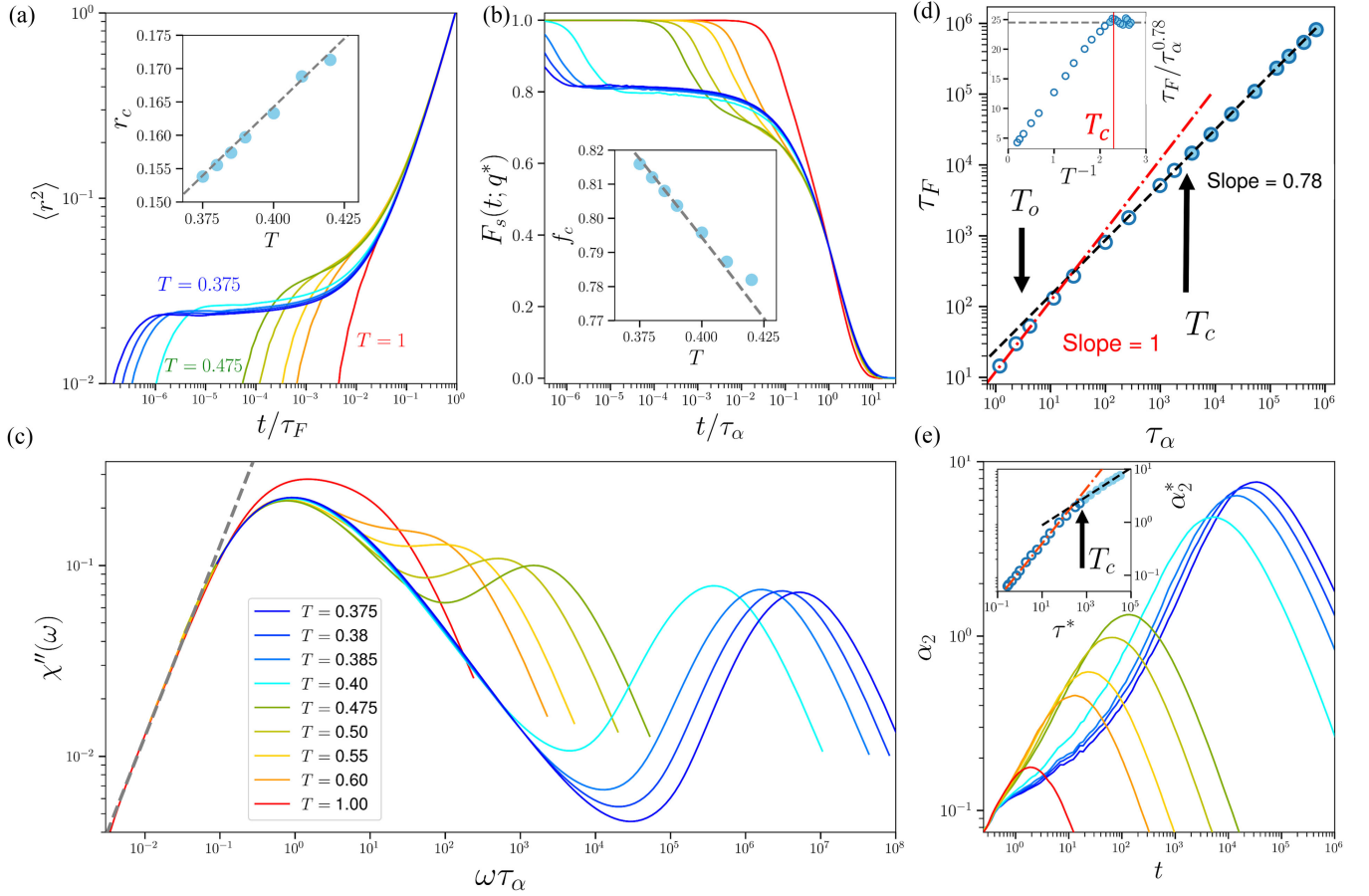


FIG. 1. Scaling laws observed in time-correlation functions. (a) MSD as a function of  $t/\tau_F$ , where  $\tau_F$  is the Fickian time, for various temperatures, as indicated in the legend reported in (c). Inset: MSD plateau height  $r_c$  as a function of temperature for  $T < T_c$ . The dashed line is a linear fit. (b) The self-intermediate scattering function (SISF) computed at wave vector  $q^* = 7.25$  (the location of the main peak in the static structure factor) as a function of  $t/\tau_\alpha$ , for the same temperatures as in (a). Inset: plateau height of the SISF  $f_c$  as a function of temperature for  $T < T_c$ . The dashed line is a fit with a Gaussian:  $f_c \propto \exp[-aT^2]$ . (c) Dynamic susceptibility  $\chi''(\omega)$  as a function of rescaled frequency  $\omega\tau_\alpha$  for the temperatures indicated in the legend. Gray dashed line is a power law  $\propto \omega$ . (d)  $\tau_F$  versus  $\tau_\alpha$  for temperatures above and below  $T_c$  (open and filled circles, respectively). The orange line is a linear fit to the data for  $T \geq T_o$  (open circles), where  $T_o \approx 0.8$  is the estimated onset temperature of glassy dynamics for this system [25,52]; the black line is a power-law fit  $\tau_F \propto \tau_\alpha^{0.78}$  to the data for  $T < T_c$ . Inset: a different representation of the data in the main panel is shown, as a function of  $1/T$  (see text). (e) Non-Gaussian parameter (NGP),  $\alpha_2 = 5\langle r^4(t) \rangle / 3\langle r^2(t) \rangle^2 - 1$ , as a function of time for the same temperatures as in (a). Inset: maximum value  $\alpha_2^*$  of the NGP as a function of its characteristic time  $\tau^*$ . Dashed lines are power-law guides to the eyes having slopes 0.52 (red line) and 0.27 (black line), describing high-temperatures (open circles) and low-temperatures (filled circles) data.

$\alpha$ -relaxation time  $\tau_\alpha$  (see SM [58]). In agreement with MCT, the curves at intermediate  $T$ 's fall onto a master curve when particles are exiting the cage, with a decay from a plateau that is well described by a power law  $f_c - B(T)t^b$ , with the nonergodicity parameter  $f_c$  and the von Schweidler exponent  $b$  that are independent from  $T$ . In contrast, for low  $T$ 's the correlators no longer follow a master curve when exiting the plateau, with  $f_c$  now rising with decreasing  $T$  because of anharmonic effects; see inset of Fig. 1(b). Note that the crossover in the  $T$  dependence of  $f_c$ , mirroring the crossover of  $r_c$ , shows that the change in transport mechanism is not only seen on the timescale of the  $\alpha$  relaxation, but also on the scale of the vibrations, which opens the possibility to detect this crossover in

various types of experiments that can access these short-time quantities. Hence, we again find qualitative differences in the dynamics at intermediate and deeply supercooled conditions (approximately, above and below  $T_c$ ). Since above  $T_c$  the size of the cage is a constant, we conclude that this size  $r_c$  shows below  $T_c$  a  $T$  dependence that is very different from the one above  $T_c$ , reflecting the qualitative change of the dynamics even on the timescales of the vibrational motion.

In experiments, it is the frequency domain that gives access to the largest dynamic range, and therefore the dynamic susceptibility  $\chi''(\omega; T)$ , i.e., the imaginary part of the time-Fourier transform of the time-correlation function  $F_s(q, t)$ , has often been used to characterize the relaxation

dynamics of glass formers [59]. In numerical simulations the measurement of  $\chi''(\omega; T)$  is unfortunately not straightforward, since the signal can be very small and is hence affected by noise [39,47,60]. Consequently, to date, the investigation of  $\chi''$  in simulations of low-temperature systems has remained largely unexplored, despite the utility of this function to identify scaling laws in the dynamics. One of the most intriguing scaling is the  $\omega^{-\gamma}$  decay of  $\chi''$  on the high-frequency side of the  $\alpha$  peak, a  $\omega$  dependence found in a multitude of glass formers at low temperatures (with  $\gamma \approx 0.5$ ), for which there is so far no solid microscopic interpretation [51,61]; below, we will demonstrate that this phenomenon is a direct consequence of the relaxation mechanism that becomes dominant at  $T$ 's below  $T_c$ .

In Fig. 1(c) we present  $\chi''(\omega\tau_\alpha; T)$  as computed directly from the trajectories, i.e., without any postprocess smoothing of the curves (see Sec. IV for details). At high temperatures,  $T > T_c$ ,  $\chi''(\omega\tau_\alpha; T)$  displays a single, nearly symmetric peak associated with the quasiexponential relaxation of the SISF, as expected. Upon moderate supercooling, the spectrum progressively broadens and features two peaks: a microscopic peak at high frequencies, related to vibrational dynamics, and a prominent peak at low frequencies, the  $\alpha$  peak located at  $\omega\tau_\alpha \approx 1$  associated with structural relaxation. The  $\alpha$  peak becomes increasingly asymmetric as temperature decreases, reflecting the well-known progressive stretching of the decay of the SISF [24,33,62], discussed in detail in the context of Fig. 2. In the deeply supercooled regime, the two peaks become well separated (by almost seven decades in frequency at the lowest  $T$ ), while their heights and amplitudes saturate. This graph demonstrates that at these  $T$ 's the  $\alpha$  peak becomes independent of  $T$  and that this scaling does not only hold in the low-frequency regime,  $\omega\tau_\alpha \lesssim 1$ , but also for frequencies several decades above the  $\alpha$  peak. Note that the presence of this master curve is not the result of fixing the shape of the  $\alpha$  peak at low temperatures as it has been done, e.g., in Ref. [39], but reflects the genuine  $\omega$  dependence of the raw data, a result that to the best of our knowledge is a first for numerical simulations of a glass former below  $T_c$ . Our observations will be shown below to agree perfectly with our finding of the SISF that shows a scaling behavior at very low temperatures [see Fig. 2(b) and our discussion of Fig. 2 below].

Figure 1(d) shows the relation between the two time-scales  $\tau_\alpha$  and  $\tau_F$ . For high and intermediate temperatures one finds the expected Stokes-Einstein relation, i.e.,  $\tau_F \propto \tau_\alpha$ . Slightly above  $T_c$  this dependence is replaced by a fractional Stokes-Einstein relation  $\tau_F \propto \tau_\alpha^c$  [24,63], with an exponent  $c$  around 0.78, a value that is close to the one found in Refs. [24,38] in similar analyses, indication of strong dynamical heterogeneities (DH) in the sample [64,65]. In the inset of Fig. 1(d) we present  $\tau_F/\tau_\alpha^{0.78}$ , and one recognizes that the crossover between the two regimes occurs slightly above  $T_c$ .

One well-known indicator for the strength of these DHs is the non-Gaussian parameter [66,67]  $\alpha_2(t) = 5\langle r^4(t) \rangle / 3\langle r^2(t) \rangle^2 - 1$ , the  $t$  dependence of which is presented in Fig. 1(e). One recognizes that at high and intermediate temperatures the rise of  $\alpha_2(t)$  is essentially independent of  $T$  and is well described by a power law, in agreement with earlier studies [52]. For temperatures below  $T_c$ , this is no longer the case, showing that there is a change in the nature of the  $T$  dependence of the DH. (The  $T$  dependence of  $\tau^*$ , the time at which  $\alpha_2$  reaches its maximum, is presented in Fig. S5 of SM [58].) The inset of Fig. 1(e) shows  $\alpha_2^* = \alpha_2(\tau^*)$  as a function of  $\tau^*$ . One clearly sees that there are two regimes, both of which are described well by a power law, but with an exponent that depends on the regime, in qualitative agreement with the prediction of Ref. [21]. The crossover point between these two regimes is close to  $T_c$ , a further evidence that at this temperature the nature of the dynamics changes.

Overall, Fig. 1 demonstrates that, upon changing  $T$ , there are two different dynamic regimes, each of which has its own scaling laws: For  $T$  above  $T_c$  one has the behavior predicted by MCT, while for temperature below  $T_c$  we reveal the existence of laws that to the best of our knowledge have not been identified so far. We do not claim, however, that  $T_c$  is the exact boundary between these two different dynamical behaviors. Rather, this temperature should be regarded as indicative of a point at which the nature of the dynamics changes, and we emphasize that we reach this conclusion directly from the analysis of the raw data, i.e., without making any MCT-inspired fit.

## B. Emergence of subdiffusive scaling laws in the deeply supercooled regime

In the following we focus on the relaxation dynamics at low  $T$ 's and demonstrate that it shows system-universal features that permit us to describe this dynamics in a simple manner. Figure 2(a) shows that the escape from the cage, characterized by the difference  $\langle r^2(t; T) \rangle - r_c^2(T)$ , is a process that is independent of  $T$  when monitored as a function of rescaled time  $t/\tau_F$ . For times shorter than  $\tau^*$ , this difference is accurately described by a power law with an exponent  $b' = 0.66 \pm 0.05 \approx 2/3$ . For times larger than  $\tau^*$ , the curve starts to bend upward and enters the Fickian regime around  $\tau_F$  [53,68]. To the best of our knowledge, the presence of such a unique subdiffusive behavior at the end of the caging regime, at temperatures significantly below  $T_c$ , has never been observed in earlier studies of supercooled liquids because the data were not sufficiently precise. Recent experiments for molecular systems by Caporaletti *et al* do, however, show a  $t$  dependence that is compatible with such a power law [69], indicating that the results from our simulations reflect the dynamics of real glass formers.

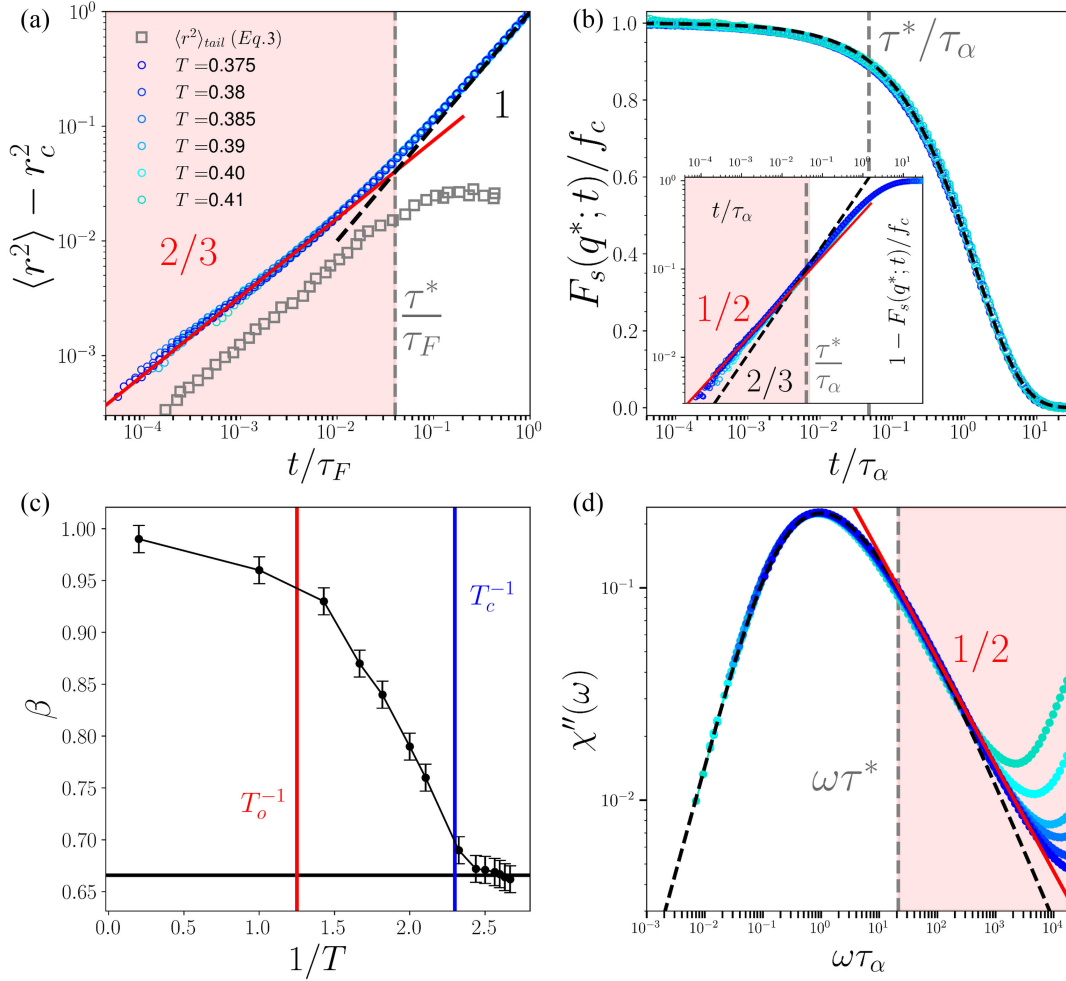


FIG. 2. Power laws in the short-time dynamics and stretching of the  $\alpha$  relaxation. (a)  $\langle r^2(t) \rangle - r_c^2(T)$  as a function of  $t/\tau_F$ , for different temperatures in the sub- $T_c$  regime. Black and red lines are  $t$  and  $t^{2/3}$  power laws, respectively. Contribution  $\langle r^2(t) \rangle_{\text{tail}}$  (see text) for  $T = 0.375$  is plotted as gray points. (b) The colored curves show the SISFs rescaled by the nonergodicity factor  $f_c(q^*)$  as a function of  $t/\tau_\alpha$ , for the same temperatures as in (a). The black dashed line is a fit with a stretched exponential with exponent  $\beta = 2/3$ . Inset: the black dashed line indicates the short-time expansion  $\propto t^\beta$  of the stretched-exponential fit, red dashed line is a fit at short times representing  $t^{0.5}$ . (c) Stretching exponent  $\beta$ , as obtained from the intermediate scattering function, as a function of inverse temperature, at various temperatures. The red vertical line indicates the critical temperature  $T_c = 0.435$ ; black horizontal line indicates the value  $\beta = 2/3$ . (d) Dynamic susceptibility  $\chi''$  as a function of rescaled frequency  $\omega\tau_\alpha$ , for the same temperatures in (a), around the  $\alpha$ -peak region. The black dashed line is obtained for a stretched-exponential decay of the SISF with  $\beta = 2/3$ . The red dashed line is a  $\propto \omega^{0.5}$  fit to the high-frequency data. Vertical dashed gray lines in the panels indicate the position of  $\tau^*/\tau_\alpha$  at the lowest temperature.

That the escape process from the cage is independent of  $T$  can also be inferred from  $F_s(q^*, t)$  in that for temperatures below  $T_c$  the ratio  $F_s(q^*, t; T)/f_c(T)$ , Fig. 2(b), falls indeed on a master curve when plotted as a function of  $t/\tau_\alpha$ . This master curve is very well described by a stretched exponential,  $\exp\{-(t/\tau_\alpha)^\beta\}$ , with a  $T$ -independent  $\beta = 2/3$ . To the best of our knowledge, the identification of such a clear-cut master curve for the  $\alpha$  relaxation in the time domain has never been reported before for glass-forming liquids. This  $T$  independence of  $\beta$  at low  $T$  can be appreciated in Fig. 2(c) where we show  $\beta(T)$  as obtained from our data for the full range of investigated temperatures,  $5.0 \geq T \geq 0.375$  (see Sec. IV for details on the

extraction of  $\beta$ ). It is evident that at high and intermediate  $T$ 's,  $\beta$  depends significantly on  $T$ , but that at the lowest  $T$ 's it saturates, confirming that, for deeply supercooled conditions, a new dynamic regime is approached.

Returning to Fig. 2(b), one notes for short times,  $t \lesssim 0.01\tau_\alpha$ , the stretched-exponential fit does not describe the data very accurately. To examine this deviation we present in the inset the difference  $1 - F_s(t/\tau_\alpha)/f_c$  in a double-logarithmic plot and one recognizes that the exit from the plateau is captured well by a power law  $t^{1/2}$ , up to times around  $\tau^*$ , at which point the stretched-exponential decay sets in. Thus the short-time dependence is not given by a Taylor expansion of the stretched exponential

(dashed black line), in qualitative agreement with the findings at higher temperatures, where the late  $\beta$ -relaxation exponent  $b$  and the stretching exponent  $\beta$  of the  $\alpha$  relaxation generally do not coincide [48]. Note that the  $t^{1/2}$  power-law behavior emerges precisely in the time window in which the MSD exhibits its characteristic  $t^{2/3}$  scaling [Fig. 2(a)], indicating that both phenomena are intimately connected to the distinctive mechanism by which particles escape their cages at very low temperatures. Below we will come back to this power-law dependence.

Figure 2(d) shows  $\chi''(\omega\tau_\alpha; T)$  from Fig. 1(c), restricted to frequencies around the  $\alpha$  peak and to temperatures well below  $T_c$ . All data collapse onto a master curve, both in the low-frequency regime and up to several decades above  $\omega\tau_\alpha = 1$ . The dashed line in the graph is the Fourier transform of the stretched-exponential fit shown in Fig. 2(b), with the fixed stretching exponent  $\beta = 2/3$ , and it perfectly captures the shape of the master curve up to frequencies around  $100\omega\tau_\alpha$ . For still higher frequencies, clear deviations emerge, and these are not related to the presence of the minimum between the  $\alpha$  peak and the vibrational peak since the data for different  $T$ 's follow the master curve even at  $\omega$  just slightly smaller than the location of the minimum. Indeed, these deviations are themselves temperature independent, and are accurately described by a power-law master decay  $\chi'' \sim (\omega\tau_\alpha)^\gamma$ , with  $\gamma = -1/2$ . Note, however, that this  $-1/2$  power-law behavior gives a good description of the master curve already for frequencies  $\omega\tau_\alpha \geq 20 \approx \omega\tau^*$ ; i.e., there is approximately one decade  $[\omega\tau^*, \omega\tau_\alpha]$  in which stretched-exponential and power-law fits of the master curve are hardly distinguishable, even with our high-quality data.

High-frequency power laws in  $\chi''$ , often with an exponent close to  $-1/2$ , have frequently been observed in glass-forming systems [51], suggesting that this dependence represents a universal feature of glassy dynamics. This “excess contribution” to the  $\alpha$  peak, whose microscopic origin is still a matter of intense debate [51,61], is frequently referred to as *excess wing*. Some authors use this term to denote an underlying short-time contribution to  $\chi''(\omega; T)$ , possibly stemming from the motion of internal degrees of freedom, such as a back-and-forth motion of an end group in a molecule, while others connect it with dynamic facilitation or spatial heterogeneous dynamics [39,51,59,61,70–76]. To avoid any confusion, hereafter we operationally define excess wing as the portion of the  $\chi''(\omega\tau_\alpha)$  master curve at frequencies  $\omega\tau^* > 1$  that can be accurately described by a power law, i.e., that is in excess of the stretched exponential.

Inspection of the data in the inset of Figs. 2(b) and in 2(d) shows that the two power laws are seen in the same dynamical window. This hints that the excess wing and the power-law decay from the plateau in the SISF are closely related to each other, and below we will demonstrate that

this behavior is directly related to *rare cage-escaping events* of the particles. To the best of our knowledge, the simultaneous identification of such well-defined power-law master scalings for the MSD, SISF, and  $\chi''$  in the postcaging regime up to  $t = \tau^*$  has not been reported before. Below we will derive an analytical expression for the reported subdiffusive scalings and show that the observed power laws are only effective ones, while the full time dependence is more complex. Finally, we stress that the data collapse found for the various dynamical correlators in Fig. 2 start to be observable at the end of the caging regime, but persist also at very long times, demonstrating that the dynamics at deeply supercooled conditions is simple, i.e., fully independent of temperature upon proper rescaling of time.

### C. Characterizing the cage-escape via the Van Hove function

The results of Figs. 1 and 2 imply that the properties of the subdiffusive dynamics at the end of the caging regime are intimately related to non-Gaussian deviations of the particle displacement distribution, which are most pronounced at  $t = \tau^*$ . To obtain deeper insight into the nature of the observed low- $T$  dynamics, we thus probe the self-Van Hove function  $G_s(r, t)$  which is related to the probability that a particle has moved in time  $t$  by a distance  $r$ . Figure 3(a) shows that, at short times,  $4\pi r^2 G_s(r, t)$  peaks at a distance  $r_1$ , i.e., it has a Gaussian-like core due to the particles that rattle inside the cage they occupied at  $t = 0$ , and for a long time (up to around  $\tau_F$ ; see below) the peak position remains basically fixed at  $r_1 \approx r_c$ . For times around  $\tau^*$  the distribution shows a shoulder which evolves into a second peak at a distance  $r_2 \approx 0.9$ , i.e., close to the position of the main peak in the radial distribution function [46]. This second peak is ascribed to the particles that have left their original cage and have effectively hopped to the location that corresponds to the first neighbor distance [33,39]. This second maximum becomes predominant at long times ( $t > \tau_F$ ) and subsequently starts to move to larger distances, while the first peak decreases and eventually fades away, indicating that the dynamics has become diffusive since all the particles have left their original cage.

Remarkably, the distribution displays for all times an exponential-like tail, compatible with the prediction of Ref. [22], present even at very short times ( $t = 0.01\tau^*$ ), i.e., the original cages start to “leak” very early, and this is the reason that the MSD detaches from the plateau. This exponential tail persists even at very long times ( $t > \tau_F$ ), where Fickian non-Gaussian diffusion [53,77,78] is at play [ $\alpha_2$  has not yet decayed to zero; see Fig. 1(e)]. For temperatures above  $T_c$ , Chauduri *et al.* have found that exponential tails are present in a variety of glass formers [79], and thus our finding generalizes these results to the  $T$

range below  $T_c$ . However, the observation that around  $\tau^*$  this tail starts at a distance  $r \approx 1$  indicates that at low temperatures the effective hopping distance is given by the nearest neighbor distance, in contrast to the finding of Ref. [79] for which a roughly 3 times smaller distance was found for  $T > T_c$ , a value that is compatible with the conclusions reached by the direct analysis of the cage jumps within single-particle trajectories [80] and with the predictions of Ref. [21]. (Representative examples of the trajectories at  $T = 0.475$  and  $T = 0.380$  are presented in the SM [58] and one sees that their qualitative nature depends indeed on whether  $T$  is above or below  $T_c$ .) Thus, this is further evidence that the dynamics above  $T_c$  differs qualitatively from the one below  $T_c$ . Figure 3(b) illustrates the qualitative differences in the shape of  $G_s(r, t)$  for temperatures above or below  $T_c$  at fixed  $t = \tau^*(T)$ . At intermediate temperatures,  $T = 0.55$ , the distribution shows a single maximum and no sign of a shoulder is visible; i.e., the caging core is broad and connects smoothly to the exponential tail. As temperature is lowered significantly below  $T_c$ , the width of this core shrinks, and a pronounced shoulder is present before the exponential tails. We note that at low  $T$ ,  $G_s(r, t)$  at  $t = \tau^*$  seems to converge to a  $T$ -independent master curve, hinting that the beginning of the  $\alpha$ -relaxation process tends to become independent from  $T$  (if measured on a rescaled timescale) and hence “simple,” coherent with the findings of Figs. 1 and 2.

Figures 3(a) and 3(b) also demonstrate that below  $T_c$  the dynamics is much more heterogeneous than above  $T_c$  in that, at the lowest temperatures and up to a time of order  $\tau_F$ , there are two populations of particles that are clearly distinct: those that are still caged and those that have escaped from their original cages. This is in contrast to

the case of higher temperatures for which this difference is much less pronounced. For intermediate  $T$ 's, the displacement far away from the cage is due to a sequence of rare (short) jumps of length around 0.3 [79]. Instead, at very low temperatures, we observe a few rare events in which particles hop by a distance of  $O(1)$ , before vibrating again in a new cage at  $r_2$ . [For the lowest  $T$  one indeed sees in Fig. 3(b) a further weak peak at  $r \approx 2$  due to particles that have jumped twice.] In both cases, exponential tails are the result of successions of rare events. The relative importance of the two peaks is quantified in Fig. 3(c) where we plot the  $t$  dependence of their height for the lowest temperatures. The peak at  $r = r_1$  is the only one present at very short times, and its magnitude remains basically constant for  $t \lesssim \tau^*$ , i.e., in the time- in which one observes the  $2/3$  power-law growth in the MSD discussed above. In this time range, no secondary maximum exists, but we still can read off the value of  $G_s(r, t)$  at  $r_2 = 0.9$ , and one finds that it grows approximately with a  $2/3$  power law (red dashed line), a result that establishes the connection between  $G_s(r_2, t)$  and the MSD that shows the same time dependence. For larger times,  $\tau^* \leq t \leq \tau_F$ , the shoulder at  $r_2$  becomes a real peak and reaches its maximum height, before decreasing at very long times. Only for times well beyond the Fickian onset this secondary peak becomes the highest one, which demonstrates that Fickian diffusion does not imply that all particles have left their original cage. In conclusion, the subdiffusive growth of the MSD is due to particles that have hopped once (secondary maximum) and, to a lesser extent, those anomalously far from their original cages (exponential tails). The fact that in this low- $T$  regime this simple short-time dynamics depends only on  $t/\tau_F$  but not on  $T$  is highly nontrivial, indicating a *distinctive*

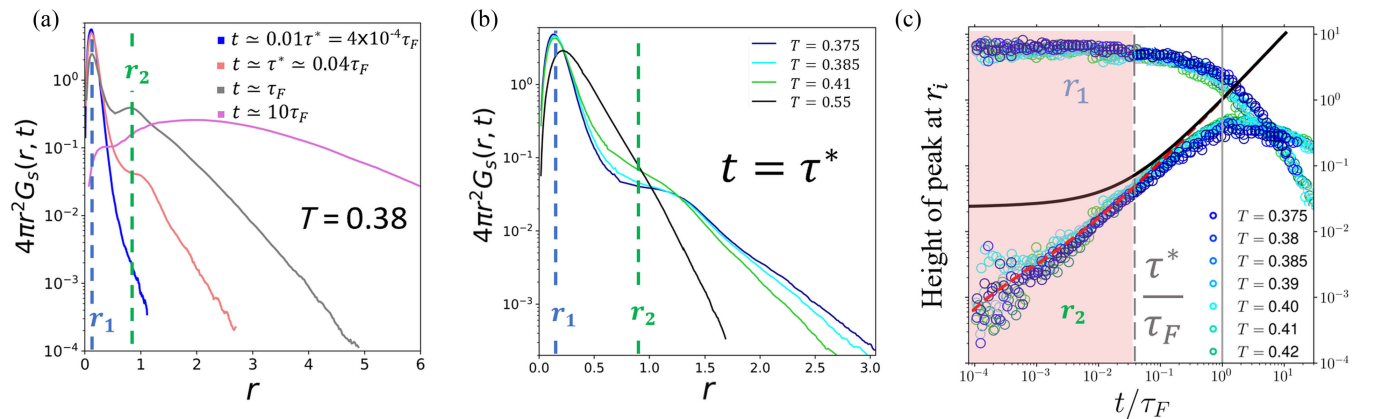


FIG. 3. The time and temperature dependence of the Van Hove function. (a)  $r$  dependence of  $4\pi r^2 G_s(r, t)$  at  $T = 0.38$  for different times, indicated in the legend. (b)  $4\pi r^2 G_s(r, t)$  as a function of  $r$ , for different temperatures  $T$ , indicated in the legend, at  $t = \tau^*(T)$ . The vertical dashed lines show the length scales  $r_1$  and  $r_2$ , the size of the cage and the hopping distance, respectively. (c) Value of  $4\pi r^2 G_s(r, t)$  at  $r = r_1$  and  $r = r_2$ , as a function of rescaled time. The temperatures are the same as in (b).  $\langle r^2(t) \rangle - r_c^2$  is plotted as red dashed line,  $\langle r^2(t) \rangle$  is plotted as black solid line, for  $T = 0.375$ . Vertical dashed line indicates the location of  $\tau^*$  at the lowest temperature, vertical solid line indicates the location of  $\tau_F$ .

*uncaging process* in which the dynamics shows a power-law dependence on time with an exponent that is independent of temperature even if the cage size depends (in a nonharmonic way) on  $T$ .

To clarify further the link between the power-law dependences documented in Fig. 2 and the exponential tails in  $G_s(r, t)$ , we present in Fig. 4(a) the one-dimensional Van Hove function at the lowest  $T$ . To compare the distribution at different times, we plot this probability  $P(X, t)$  as a function of the reduced variable  $X = x/\sqrt{\langle x^2(t) \rangle}$ . For a Gaussian process, this rescaling results thus in a universal time-independent master curve:  $G(X) = \sqrt{2/\pi} \exp[-X^2/2]$ , included in the figure as well. For large  $X$  all distributions shown in Fig. 4(a) display an excess probability with respect to  $G(X)$ . Note that in this representation, the peak at  $r_2$  seen in Fig. 3(a) is not visible anymore, but is replaced by a marked shoulder for times

around  $\tau^*$ , where deviations from  $G(X)$  are most pronounced. For large displacements, i.e.,  $X \geq 6 \stackrel{\text{def}}{=} X_0$ , which is beyond the shoulder at  $r_2$ , the tails are well described by an exponential  $A(t) \exp[-(X - X_0)/L(t)]$ . [Note that, at  $t = \tau^*$ , the condition  $X > X_0$  corresponds for the 3D  $G_s(r, t)$  to the condition  $r > r_2$ , since  $6\langle r^2(\tau^*) \rangle \approx 6r_c^2 \approx 0.9$ .] One sees immediately that the amplitude  $A(t)$  shows a strong dependence on time, while the decay length  $L(t)$  depends only weakly on  $t$ , see Figs. 4(b) and 4(c).  $A(t)$  increases by more than one order of magnitude for  $t < \tau^*$  and peaks at  $t \simeq \tau^*$ . Remarkably, the curves for the different  $T$ 's fall onto the same master curve before  $\tau^*$ , showing a power law with an exponent  $\approx 0.5$ . We note that for temperatures above  $T_c$  no such master curve is observed (see Fig. S6 [58]). Two further features of  $A(t)$  for  $t > \tau^*$  are not found at higher temperatures, namely, the presence of a shoulder (at around  $t = 8\tau^*$ ) and the  $T$  independence of

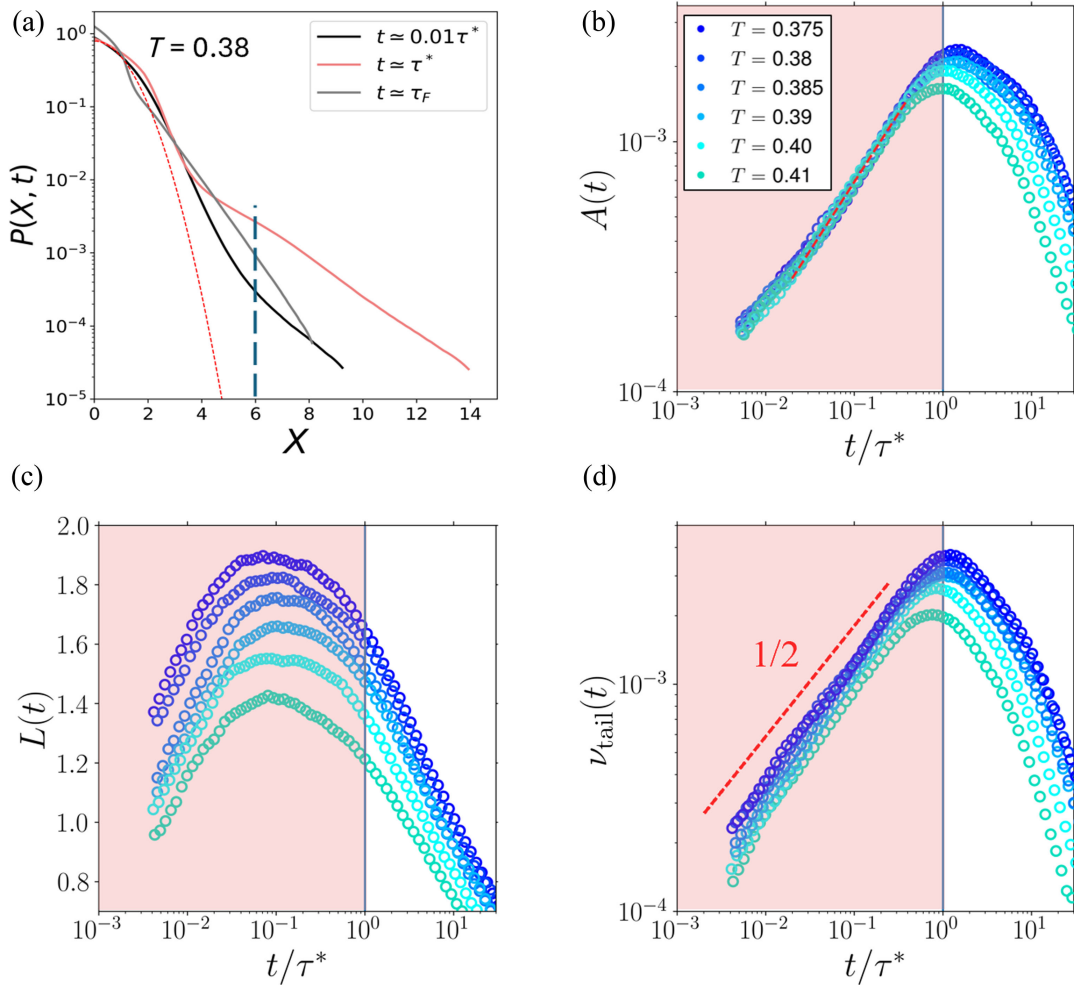


FIG. 4. Characterizing the dynamics of the leaked particles. (a) Normalized 1D displacement distribution function as a function of scaled distance  $X$  for different times at  $T = 0.38$ . The vertical dashed line is located at  $X = X_0$ , which corresponds to the hopping distance of the particles at  $t = \tau^*$ . The red dashed line is a normalized Gaussian. Panels (b) and (c) show the parameters of the exponential fit to the tails, as a function of  $t/\tau^*$  for the lowest temperatures. (d) The fraction of particles that have leaked outside the Gaussian core as a function of  $t/\tau^*$ .

the shape of the curve when it decays; see Fig. S6 in SM [58].

Also  $L(t)$  shows a nonmonotonic behavior, Fig. 4(c), with a  $t$  dependence that is similar for all temperatures, and has a maximum approximately at  $t = 0.1\tau^*$ . Throughout the examined  $t$  range, however,  $L(t)$  at fixed  $T$  varies at most by 35% of its maximum value. Thus one concludes that it is the amplitude of the tails, rather than their extent, which is the key ingredient for the DH. [In Fig. S7 we report the time evolution of the dimensional decay length  $l(t) = L(t)\sqrt{\langle x^2(t) \rangle}$  [58], and one sees that the variation with time is not very pronounced; i.e.,  $L(t)$  reflects indeed the relevant length scale for the dynamics.] In Fig. 4(d) we present  $\nu_{\text{tail}}(t)$ , the fraction of particles that have made an  $X$  displacement larger than  $X_0$ , computed analytically as the total area  $A(t)L(t)$  under the tail of the distribution function. We recognize that up to times  $\tau^*(T)$  this fraction shows a clear power-law behavior, with an exponent ( $\approx 0.5$ ) that is nearly independent of  $T$  and a prefactor that depends only mildly on  $T$  via the  $T$  dependence of  $L$  shown in Fig. 4(c).

These observations demonstrate that we are here probing a  $T$  regime in which the dynamics is qualitatively different from the one at or above  $T_c$ , and becomes qualitatively independent of  $T$ , if time is appropriately rescaled. Up to  $\tau^*$ , the evolution of  $P(X, t)$ , and hence  $G_s(r, t)$ , is given by the superposition of two processes: a decrease of the height of the Gaussian core and a related increase of the exponential tail, in an almost rigid fashion with only a slight broadening.

In the next section we will show how the time dependencies of  $\nu_{\text{tail}}$  and  $L(t)$  dictate the subdiffusive behaviors shown in Fig. 2, and specifically how the  $1/2$  power law of  $\nu_{\text{tail}}$  is mirrored in the power-law behaviors of  $F_s$  and  $\chi''$ , at times below  $\tau^*$ .

#### D. Connecting rare-escaping events and subdiffusive scaling

The fact that at low  $T$  the Van Hove function in 1D can be approximated well by the sum of a Gaussian and an exponential tail can be exploited to describe the Van Hove function in 3D, in particular for times  $t < \tau^*$ , of interest in this section. The 3D Van Hove function is given by

$$4\pi G_s(r, t)r^2 = \begin{cases} \alpha'(t)r^2 e^{-r^2/2\sigma^2} & \text{if } r < r_0 \\ \alpha''(t)r^2 e^{-r/L(t)} & \text{if } r \geq r_0, \end{cases} \quad (2)$$

where  $r_0$  is the lower boundary for the exponential tail. The functions  $\alpha'(t)$  and  $\alpha''(t)$  are related to the relative weights between caged and cage-escaping particles, and can be determined from the exponential decay of the 1D distribution and a normalization condition; see details in SM [58].

For  $\langle r^2(t) \rangle_{\text{tail}}$ , i.e., the contribution to the MSD from the particles that are in the exponential tail of  $G_s(r, t)$ , it is found that (see Appendix A)

$$\langle r^2(t) \rangle_{\text{tail}} = r_c^2 X_0^2 \nu_{\text{tail}}(t) \frac{[1 + \phi(t)]}{1 - \nu_{\text{tail}}(t) X_0^2 [1 + \phi(t)]}, \quad (3)$$

where  $\phi(t) = 2L(t)/X_0 + 2L^2(t)/X_0^2$ . Thus the time dependence of  $\langle r^2(t) \rangle_{\text{tail}}$  is given by  $\nu_{\text{tail}}(t) \propto t^{0.5}$  [Fig. 4(d)], enhanced by the second factor on the right-hand side of Eq. (3), resulting in the effective power law with exponent  $\approx 0.67$  shown in Fig. 2(a). (See Fig. S11 for a quantitative demonstration [58].) In Fig. 2(a) we have also included  $\langle r^2(t) \rangle_{\text{tail}}$ , for the sake of clarity only for  $T = 0.375$ , and one sees that its time dependence does indeed track the one of  $\langle r^2(t) \rangle$ . [That the prefactor is not quite the same is likely related to the approximation made in Eq. (2), including the ensuing choice of the cutoff for the exponential tail.] Similar results hold for the other temperatures below  $T_c$ .

Equation (2) for  $G_s(r, t)$  allows us to obtain also an expression for the SISF (see Appendix B), which for times  $t \lesssim \tau^*$  is in leading order given by

$$F_s(t; q^*) = 4\pi \int G_s(r, t) \frac{\sin(q^*r)}{q^*r} r^2 dr \simeq f_c(q^*) - k(q^*)\nu_{\text{tail}}(t). \quad (4)$$

Since  $\nu_{\text{tail}}(t) \propto t^{1/2}$ , this result explains the power law, with the same exponent, identified for  $F_s(t)$  in the inset of Fig. 2(b).

This result for  $F_s(t)$  for  $t < \tau^*$  can be directly used to predict the  $\omega$  dependence of  $\chi''(\omega)$  in the corresponding frequency range, i.e.,  $\omega > 1/\tau^*$  (neglecting the very short-time dynamics described by the susceptibility at high frequency):

$$\chi''(\omega > 1/\tau^*) = - \int \frac{dF_s(q^*, t)}{dt} \frac{\omega t}{1 + (\omega t)^2} dt \propto \omega^{-1/2}. \quad (5)$$

Equations (3)–(5) reveal the direct connection between the term  $\nu_{\text{tail}}(t)$  and the dynamic correlation functions  $\langle r^2(t) \rangle$ ,  $F_s(q, t)$ , and  $\chi''(q, \omega)$  for the time window in which the particles start to leave their cage. The scaling laws documented in Fig. 2 are thus a direct consequence of the power-law dependence of  $\nu_{\text{tail}}(t)$  on time, i.e., the time dependence of the number of particles having undergone a rare cage-escaping event.

Here, the question naturally arises: Why are the subdiffusive exponents for the MSD and for the SISF different, see Fig. 2, at variance with the MCT result for the von Schweidler regime? This can be intuitively understood. Unlike  $F_s$ , whose time dependence is proportional to  $\nu_{\text{tail}} \propto t^{1/2}$ , the MSD is significantly affected also by the

time dependence of the  $\phi(t)$  term, which itself is determined by the length scale  $L(t)$  of the exponential tail (see Fig. S11 [58]). This happens because the MSD is an  $r^2$ -weighted average of  $G_s(r, t)$ , and is therefore more sensitive to large  $r$  displacement; as a result, even a slight broadening of the exponential tail, while negligible for other observables, turns out to be in fact amplified for the MSD. In short, while the departure from the plateau in the SISF is primarily governed by *how many* particles escape their cages (i.e., enter the tails of the Van Hove function), the MSD is also sensitive to *how far* these particles move once they have escaped. This argument clarifies why, in the deeply supercooled regime—where the exponential tails are more pronounced and also hopping dynamics is at play—the MSD and  $F_s$  depart from their plateaus with different exponents, whereas in the framework of MCT (as confirmed by simulations at higher temperatures) they typically exhibit the same scaling, with a unique exponent close to  $1/2$ . This difference in exponents is hence a specific feature of the asymptotically low- $T$  dynamics and a further (one-particle) facet of increasing DH.

### III. DISCUSSION

In this work we have probed the details of the relaxation dynamics below  $T_c$ . Emphasis was given to confront this dynamics with the one found at temperatures above  $T_c$ , focusing not only on the short timescales as has been done in previous studies, but probing also the  $\alpha$  process, which thus requires us to access particularly long timescales. Although the swap Monte Carlo algorithm allows us to equilibrate some systems even at very low temperatures, the characterization of the realistic relaxation dynamics requires the use of standard molecular dynamics simulations as used here, thus limiting the  $T$  range at which the  $\alpha$ -relaxation decay can be followed completely. Our study of the relaxation dynamics at temperatures well below  $T_c$  reveals that this dynamics shows unexpected features, some of which render the description of the dynamics more complex, while others simplify it. The fact that the size of the cage decreases linearly with  $T$  [see insets of Figs. 1(a) and 1(b)], i.e., that the cage is strongly anharmonic [33], is surprising since the MSD shows that below  $T_c$  the particles are trapped in their cage for times spanning several decades of their oscillation period, indicating that the cage is very stable. This anharmonicity, setting in at temperatures somewhat below  $T_c$ , will influence the  $T$  dependence of the specific heat and is thus an experimentally accessible quantity [81,82]. The marked presence of these anharmonic effects also implies that the prediction of MCT that at  $T_c$  the height of the plateau shows a square-root singularity [9] is not correct for the present system, and we deem that this discrepancy, predicted in Ref. [21], may not be a particularity of the glass former we studied, but is instead a quite general result. The nature of the particle motion that

corresponds to these anharmonic vibrations is at present an open question that should be addressed in the future.

The main finding of this work is that around  $T_c$  the nature of the relaxation dynamics changes qualitatively, an insight that has been expected on general arguments [26,27,32,33,83], but has never been probed and quantified in detail. While previous studies have demonstrated that the dynamics above  $T_c$  is described well by MCT [9,46–48], we show here that below  $T_c$  it displays features that are absent at intermediate and high  $T$ 's and that contradict the predictions of the idealized version of MCT. In particular, we find that at low  $T$  the process that allows the particles to leave their cages is independent of temperature, if time is measured in terms of the Fickian time  $\tau_F$ . The Van Hove function reveals that this process is closely related to rare events in which the particles hop by a distance that is close to the nearest neighbor distance, in contrast to the case of higher  $T$ 's, for which the jump length is significantly smaller. (Although the jump length increases with decreasing  $T$ , the strongly reduced frequency of this motion makes that the relaxation dynamics does slow down with temperature.) As a consequence, these rare cage-escaping events give rise to a power-law dependence of the MSD below  $\tau^*$ , i.e., short (rescaled) times, with an exponent that is identical to the stretching exponent found in the  $\alpha$ -relaxation process, thus at long times. This equality shows that at low  $T$ 's the  $\beta$  process is linked to the  $\alpha$  process in a way that is much more direct than at  $T$ 's above  $T_c$ . Hence, the rich and complex relaxation dynamics predicted by MCT above  $T_c$  is replaced by a significantly simpler escape problem in which cooperative effects are likely less important.

The fact that, at low  $T$ , one can clearly identify a hopping length that is given by the nearest neighbor distance indicates that the relaxation process is directly related to a simple structural length scale of the liquid. This makes one expect that the results presented here are generic, i.e., do not depend on the details of the glass former considered. Indeed, one can expect that, once the temperature is significantly below  $T_c$ , the relaxation dynamics will display a distinct component of a simple activated process [84] with an activation energy that depends on the height of the free-energy barrier that needs to be crossed to move to a nearest neighbor distance [85], i.e., a short-range property of the system. This conclusion is supported by the experimental observation that the temperature dependence of the  $\alpha$  relaxation time is independent of the glass former for temperatures below  $T_c$ , once the fragility of the glass former has been scaled out [86–88]. On the other hand, close to  $T_c$ , the complex nonlinear dynamics as described by MCT makes that the  $T$  dependence of the relaxation time depends on the details of the interactions, thus determining the fragility of the glass former [89]. The connection between the here identified processes involving large particle displacement, resulting in a characteristic  $t^{0.5}$

dependence for  $\nu_{\text{tail}}(t)$ , and the shape of the high-frequency wing of the dynamics susceptibility is a natural explanation for the excess wing observed in many glass formers.

Previous studies have argued that the excess wing is a direct consequence of a facilitation dynamics [39,43]. That conclusion was reached by probing, at temperatures significantly below  $T_c$ , the relaxation dynamics of a glass former that was highly polydisperse. While the nominal polydispersity in that system was 23%, thus quite large, these low temperatures could only be reached by using the swap algorithm, and as a consequence the small particles had to constitute the vast majority of the system. Because small particles are significantly more mobile than large ones, it is not surprising that dynamic facilitation [90,91] constitutes an important relaxation channel, since slow domains in the sample will be eroded away from their surface like a sand dune during a storm. However, such a strong polydispersity is not what is found in more realistic glass formers in which the majority of the particles constitute the *slow* component in the system. Hence, even if it can be expected that dynamic facilitation is a rather generic feature of glassy dynamics, its importance for a typical glass former is still an open question. This fact has implications regarding the microscopic origin of the excess wing. While in systems in which relaxation is dominated by dynamic facilitation the mechanism leading to this wing might be related to the properties of the domains of mobile particles, our analysis of the self part of the Van Hove function reveals that the dynamics of a single tagged particle has already all the necessary ingredients to give rise to an excess wing. Thus, invoking the concept of spatial dynamical heterogeneity is not needed to rationalize this wing, since this feature can be obtained directly from the power laws in time that we have identified in the escape process from the cage, i.e., the MSD and  $F_s(q, t)$ . We also emphasize that, in contrast to Refs. [39,43], our results regarding this wing, notably its exponent, are obtained directly from the trajectories of the particles without any smoothing operation, or fixing of parameters describing the time correlation function, procedures that might give rise to numerical artifacts in the spectra. This was only possible because we considered system sizes ( $N = 32\,400$ ) that were sufficiently large to avoid finite size effects [44] and, in particular, significantly larger than the ones used in Refs. [39,43] ( $N = 1200$  in 3D).

All our conclusions in this work are based on the analysis of typical liquid state observables, like  $G_s(r, t)$ . It is, however, also important to characterize the relaxation dynamics below  $T_c$  by probing directly the properties of the particle trajectories and identifying the jump processes via a temporal filtering of these trajectories [80,92–94]. Such an analysis will allow us to study the presence of memory effects in the dynamics and hence to establish a connection with the viscoelastic behavior of the

system at low  $T$ . In addition, such studies will permit us to gain a better understanding on the origin of the power-law dependence of  $\nu_{\text{tail}}$  and whether the exponent 0.5 is universal or system dependent. Since this exponent is directly related to power law found for the excess wing, and experiments suggest that the shape of this wing depends only weakly on the system [51,61], one expects that the exponent for  $\nu_{\text{tail}}$  is basically universal, but this needs to be confirmed by simulations of other systems and analytical calculations [21,22]. We also mention that in Ref. [95] a procedure was proposed that allows us to characterize the amount of cooperativity of the particle dynamics and it was found, for the case of a highly polydisperse system, that below  $T_c$  this cooperativity decreases, in qualitative agreement with the present findings on the emergence of activated hops. This is thus evidence that the results that we have presented here are generic.

Finally, we note that the quantities presented here are all one-particle observables. It is therefore important to probe how collective quantities [66,96,97], such as the intermediate scattering function  $F(q, t)$  and correlation functions allowing us to monitor the spatial dependence of the dynamic heterogeneity, depend on temperature, i.e., whether also they show a simple  $T$  dependence below  $T_c$ . Our results on the system dynamics as probed by one-particle quantities hint to the absence of a new temperature scale below  $T_c$ . However, this does not imply that there is indeed no such temperature, which would likely correspond to the Kauzmann temperature  $T_K$ , since in principle collective quantities, such as the coherent intermediate scattering function, might indeed completely freeze at a finite temperature. Despite significant theoretical and experimental efforts to find clear evidence for the existence of a  $T_K > 0$ , there is at present no consensus that such a low temperature exists in realistic glass formers [86,87,98,99]. Advancing on this question remains thus a very challenging subject for future research [100].

## IV. METHODS

### A. Details on the simulations

We consider a binary Lennard-Jones mixture with particles that we denote by  $A$  (80%) and  $B$  (20%). The interaction between particles of type  $\alpha$  and  $\beta \in \{A, B\}$  is given by

$$V_{\alpha\beta}(r) = 4\epsilon_{\alpha\beta}[(\sigma_{\alpha\beta}/r)^{12} - (\sigma_{\alpha\beta}/r)^6], \quad (6)$$

where the interaction parameters  $\sigma_{\alpha\beta}$  and  $\epsilon_{\alpha\beta}$  are given in Ref. [48]. Schröder and Dyre showed that if the  $AA$  and  $BB$  forces from this potential are shifted and cut off at  $1.5\sigma_{AA}$ , while the one for the  $AB$  interaction is at  $2.5\sigma_{AA}$ , the system is not prone to crystallization [50]. In the present work we use  $\sigma_{AA}$  and  $\epsilon_{AA}$  as units of length and energy, setting the Boltzmann constant  $k_B$  equal to 1.0.

A comparison of the static and dynamical properties of the original model with those of the modified model shows, as long as crystallization in the original model is avoided, no significant difference between the two systems.

We have simulated this system using  $N = 32\,400$  particles at a constant density of 1.200, corresponding to a box size of 30. The time step was 0.005 and the simulations were carried out at constant energy and volume. In order to improve the statistics of the results, we have done simulations that were significantly longer than the  $\alpha$ -relaxation time (for  $T \leq 0.39$  the runs extended over  $2 \times 10^9$  time steps) and have carried out 4 independent simulations of the system at the various  $T$ 's. Simulations were carried out using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [101]. The longest runs took about 600 days of single CPU time and the total amount of CPU time used to generate the presented data corresponds to 56 years of single CPU time. Analyses of the trajectories to obtain the one-particle observables presented in the work were carried out using homemade Python codes.

The results presented in this work have been obtained by focusing on the (larger)  $A$  particles, since they are in the majority and also the slow component in the system [46,47]. For the sake of completeness we report in SM the dynamics of the  $B$  particles, qualitatively very similar to that of the  $A$  particles [58].

### B. Analysis of single-particle trajectories

The MSD for particles  $A$ , referred to in the text as  $\langle r^2(t) \rangle$ , is computed as

$$\langle r^2(t) \rangle = \frac{1}{N_A} \sum_{i=1}^{N_A} \langle |\mathbf{r}_i(t) - \mathbf{r}_i(0)|^2 \rangle, \quad (7)$$

where  $\mathbf{r}_i(t)$  is the position of particle  $i$  at time  $t$ .

The plateau height of the MSD,  $r_c(T)$ , is defined as the square root of the mean value of the MSD evaluated at all times for which  $d \log \langle r^2(t) \rangle / d \log t$  is below the threshold value of 0.02, i.e., within the caging regime.

The self-intermediate scattering function for particles  $A$ ,  $F_s(\mathbf{q}, t)$ , is computed as follows:

$$F_s(q^*, t) = \frac{1}{N_A} \sum_{j=1}^{N_A} \langle \exp\{-i\mathbf{q}^* \cdot [\mathbf{r}_j(t) - \mathbf{r}_j(0)]\} \rangle, \quad (8)$$

where  $\mathbf{q}^*$  is the wave vector corresponding to the position of the main peak in the static structure factor  $S(q)$ , i.e.,  $q^* = |\mathbf{q}^*| = 7.25$ , and on the right-hand side we have averaged over all wave vectors with the same modulus  $q^*$ . The structural relaxation time  $\tau_\alpha(T)$  is defined as the time at which the SISF has decayed to  $1/e$ .

The height of the plateau of the SISF,  $f_c(T)$ , is computed as the mean value of the SISF evaluated at all times for which  $d \log \langle r^2(t) \rangle / d \log t$  is below the threshold value of

0.02. In Fig. S4 we show that the  $T$  dependence presented in the inset of Fig. 1(a) does not depend in a significant manner on the choice of this threshold [58].

To estimate the  $T$  dependence of the stretching exponent  $\beta$  of the SISF we consider the quantity  $m(t; T) = -d \log F_s(q^*, t) / dt$ , which is independent of the height of the plateau  $f_c(T)$ . Since at long times we expect the SISF to be compatible with a stretched-exponential behavior, we fit  $\log m(t; T)$  in the range  $[0.5\tau_\alpha, 5\tau_\alpha]$  with the expression  $m_\beta(t; T) = a(T) + [\beta(T) - 1] \log t$  to estimate the stretching exponent  $\beta(T)$  presented in Fig. 2(c). In Fig. S8 we show the ratio  $m(t; T) / m_{2/3}(t, T)$ , with  $m_{2/3} = a'(T) - \frac{1}{3} \log t$  being the best fit with the expression  $m_\beta$  to the  $\log m$  data if  $\beta = 2/3$  is kept constant [58]. This representation documents the approach, as temperature is decreased toward the lowest  $T$ 's, to the master-curve behavior with  $\beta = 2/3$  of the SISF's described in the main text.

The imaginary part of the dynamic susceptibility  $\chi''(\omega)$  as a function of the frequency  $\omega$  is computed from the expression

$$\chi''(\omega; T) = - \int \frac{dF_s(q^*, t)}{d \log t} \frac{\omega t}{1 + (\omega t)^2} d \log t. \quad (9)$$

This integral can be evaluated directly from our raw data for  $F_s(q^*, t)$ ; i.e., no smoothing operation is needed.

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### DATA AVAILABILITY

The data that support the findings of this article are openly available [102].

### APPENDIX A: ESTIMATING THE CONTRIBUTION TO THE MSD OF THE PARTICLES HAVING LARGE DISPLACEMENTS

The contribution  $\langle r^2(t) \rangle_{\text{tail}}$  discussed in the main text is defined as

$$\langle r^2(t) \rangle_{\text{tail}} = 3 \langle x^2(t) \rangle_{\text{tail}} = 3 \int_{x_0(t)}^{+\infty} x^2 P(x, t) dx, \quad (A1)$$

where  $x_0(t) = X_0 \sqrt{\langle x(t)^2 \rangle}$  and  $X_0 = 6$ . Note that in the definition of the 1D displacement function the density distribution is normalized such that  $\int_0^\infty P(x, t) dx = 1$ . Making the change  $x \rightarrow X$ , and using the fact that for

$X > X_0$  the distribution in  $X$  is given by an exponential, see Fig. 4(a), we obtain

$$\langle r^2(t) \rangle_{\text{tail}} = 3 \langle x^2(t) \rangle \int_{X_0}^{+\infty} X^2 A(t) \exp \left[ -\frac{X - X_0}{L(t)} \right] dX \quad (\text{A2})$$

$$= \langle r^2(t) \rangle A(t) L(t) [X_0^2 + 2X_0 L(t) + 2L^2(t)] \quad (\text{A3})$$

$$= \langle r^2(t) \rangle A(t) L(t) X_0^2 [1 + 2L(t)/X_0 + 2L^2(t)/X_0^2]. \quad (\text{A4})$$

Recalling the result that the fraction of particles in the tail of the distribution is  $\nu_{\text{tail}} = A(t)L(t)$ , we hence obtain

$$\langle r^2(t) \rangle_{\text{tail}} = \langle r^2(t) \rangle \nu_{\text{tail}}(t) X_0^2 [1 + \phi(t)], \quad (\text{A5})$$

with  $\phi(t) = 2[L(t)/X_0 + L^2(t)/X_0^2]$ , as reported in the main text. Since the time dependence of  $\langle r^2(t) \rangle$  when the particles leave the plateau is given by  $\langle r^2(t) \rangle = r_c^2 + \langle r^2(t) \rangle_{\text{tail}}$ , one thus obtains

$$\langle r^2(t) \rangle_{\text{tail}} = \frac{r_c^2 \nu_{\text{tail}}(t) X_0^2 [1 + \phi(t)]}{1 - \nu_{\text{tail}}(t) X_0^2 [1 + \phi(t)]}. \quad (\text{A6})$$

The increase of  $\langle r^2(t) \rangle$  is hence slightly faster than the power law found for  $\nu_{\text{tail}} \propto t^{0.5}$ , rationalizing the *effective* power-law dependence  $t^{0.67}$  documented in Fig. 2(a).

## APPENDIX B: THEORETICAL PREDICTIONS FOR THE POWER LAWS IN $F_s(q, t)$ AND THE EXCESS WING IN $\chi''$

Since the system is isotropic, one reasonable generalization of the 1D distribution  $P(x, t)$  to the case of 3D is, for times  $t < \tau^*$ ,

$$4\pi r^2 G_s(r, t) = \begin{cases} \alpha'(t) r^2 e^{-r^2/2\sigma^2} & \text{if } r < r_0 \\ \alpha''(t) r^2 e^{-r/L(t)} & \text{if } r \geq r_0, \end{cases} \quad (\text{B1})$$

where  $r_0$  is the lower boundary for the exponential tail and  $\sigma$  is the width of the Gaussian describing  $G_s(r, t)$  at small  $r$ , which is close to the value of  $r_c$ . The functions  $\alpha'(t)$  and  $\alpha''(t)$  are related to the relative weights between caged and cage-escaping particles, and can be determined from the exponential decay of the 1D distribution and a normalization condition.  $\alpha''(t)$  is determined from the 1D exponential tail, imposing the condition

$$\int_{r_0}^{\infty} \alpha''(t) e^{-r/L(t)} r^2 dr \stackrel{!}{=} A(t) L(t) = \nu_{\text{tail}}(t), \quad (\text{B2})$$

which gives

$$\alpha''(t) = \nu_{\text{tail}}(t) / [2L(t)^3 + 2L^2(t)r_0 + L(t)r_0^2]. \quad (\text{B3})$$

$\alpha'(t)$  can now be determined by normalizing the total distribution:

$$\begin{aligned} & \int_0^{\infty} 4\pi r^2 G_s(r, t) dr \\ &= \int_0^{r_0} \alpha'(t) e^{-r^2/2\sigma^2} r^2 dr + \int_{r_0}^{\infty} \alpha''(t) e^{-r/L(t)} r^2 dr = 1, \end{aligned} \quad (\text{B4})$$

giving

$$\nu_{\text{tail}}(t) + \int_0^{r_0} \alpha'(t) e^{-r^2/2\sigma^2} r^2 dr = 1. \quad (\text{B5})$$

One thus obtains

$$\alpha'(t) = [1 - \nu_{\text{tail}}(t)] \frac{1}{\sigma^2 \left[ \sqrt{\frac{\pi}{2}} \text{Erf} \left( \frac{r_0}{\sqrt{2}\sigma} \right) \sigma - r_0 e^{-r_0^2/2\sigma^2} \right]}. \quad (\text{B6})$$

The SISF can now be calculated from

$$F_s(q, t) = 4\pi \int G_s(r; t) \frac{\sin(qr)}{qr} r^2 dr, \quad (\text{B7})$$

and with Eq. (B1) one obtains

$$\begin{aligned} F_s(q, t) &= \alpha'(t) \int_0^{r_0} 4\pi e^{-r^2/2\sigma^2} \frac{\sin(qr)}{qr} r^2 dr \\ &+ \alpha''(t) \int_{r_0}^{\infty} 4\pi e^{-r/L(t)} \frac{\sin(qr)}{qr} r^2 dr. \end{aligned} \quad (\text{B8})$$

Using Eqs. (B3) and (B6),  $F_s(q, t)$  can thus be written in the form

$$F_s(q, t) = [1 - \nu_{\text{tail}}(t)] f_1(q; \sigma, r_0) + \nu_{\text{tail}}(t) f_2(L(t), q; r_0), \quad (\text{B9})$$

where the functions  $f_1$  and  $f_2$  are related to the integrals in Eq. (B8) and the functions  $\alpha'$  and  $\alpha''$  in Eqs. (B3) and (B6).

To leading order in time, we can neglect the  $t$  dependence of  $f_2$  and set  $f_2(L(t), q; r_0) \approx f_2^0(q; r_0)$ , giving

$$F_s(t; q) = f_1(q; \sigma, r_0) - [f_1(q; \sigma, r_0) - f_2^0(q; r_0)] \nu_{\text{tail}}(t), \quad t < \tau^*. \quad (\text{B10})$$

Since one has  $f_1(q; \sigma, r_0) > 0$  and  $f_1(q; \sigma, r_0) - f_2^0(q; r_0) > 0$ , we can rewrite Eq. (B10) as

$$F_s(t; q^*) \simeq f_c(q^*) - k(q^*)\nu_{\text{tail}}(t), \quad (\text{B11})$$

which is Eq. (4) in the main text, and gives the form of the short-time deviation from the plateau, in agreement with the findings presented in the inset of Fig. 2(b). The link with the dynamic susceptibility in the corresponding frequency range is straightforward:

$$\chi''(\omega > 1/\tau^*) \approx - \int \frac{d}{dt} [f_c(q^*) - k(q^*)\nu_{\text{tail}}(t)] \frac{\omega t}{1 + (\omega t)^2} dt \propto \omega^{-1/2}. \quad (\text{B12})$$

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