

**Relationship between dynamical heterogeneities and stretched exponential relaxation**S. I. Simdyankin<sup>1,2</sup> and Normand Mousseau<sup>2</sup><sup>1</sup>*Department of Chemistry, University of Cambridge, Lensfield Road, Cambridge CB2 1EW, United Kingdom*<sup>2</sup>*Département de Physique and Centre de Recherche en Physique et Technologie des Couches Minces, Université de Montréal, Case Postale 6128, succursale Centre-ville, Montréal, Québec, Canada, H3C 3J7*

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We identify dynamical heterogeneities as an essential prerequisite for stretched exponential relaxation in dynamically frustrated systems. This heterogeneity takes the form of ordered domains of finite but diverging lifetime for particles in atomic or molecular systems, or spin states in magnetic materials. At the onset of the dynamical heterogeneity, the distribution of time intervals spent in such domains or traps becomes stretched exponential at long times. We rigorously show that once this is the case the autocorrelation function of the renewal process formed by these time intervals is also a stretched exponential at long times.

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**I. INTRODUCTION**

Stretched exponential relaxation (SER),  $I(t) \sim \exp[-(t/\tau)^\beta]$ , describes the relaxation of pure glasses very accurately, often over very wide ranges [1]. For example, SER describes equally well stress experiments on amorphous Se, centered on  $10^3$  s, as well as spin-polarized neutron scattering experiments, centered on  $10^{-9}$  s, both with the same value of  $\beta$ , over a time range of more than  $10^{12}$ . SER also accurately describes the stress relaxation of the superconducting transition temperatures in the cuprates, probably associated with the two-gap domain structure revealed by high-resolution scanning tunneling microscopy [2]; moreover, the measured value of  $\beta$  is the predicted value,  $3/5$ , for YBCO, by far the most extensively studied cuprate, with the highest-quality samples. The agreement is such that it has recently been proposed that the SER be used as an independent measure of sample quality in these materials [3]. Phillips' review [1] gives many more examples and demonstrates the power of SER as a guide to understanding the dynamical properties of intrinsic glasses, especially network glasses. One of the most spectacular examples was the successful prediction [4] of the long- and short-preparation-time values of  $\beta$  of orthoterphenyl (OTP), as measured by multidimensional NMR. OTP is the purest organic glass former available commercially; the predicted values were 0.43 and 0.60, and the observed values were 0.42 and 0.59.

It appears that SER may arise as a result of spatial inhomogeneities that are quenched into macroscopically pure, non-phase-separated glasses on a microscopic scale as traps or sinks that appear as part of the glass-forming process. The existence of these inhomogeneities is difficult to detect experimentally, but they readily account for SER, and predict correctly the values of  $\beta$  in many experiments [1]. Dynamical heterogeneities have been seen experimentally and numerically in glasses and supercooled liquids [5,6], spin-glass models [7,8], Lennard-Jones alloy mixtures with ellipsoidal probes [9], as well as in coupled chaotic systems such as diode-resonator arrays [10]. In particular, the concept of dynamical heterogeneities has emerged as critical for the description of the microscopic dynamics associated with the dramatic slowing down of the relaxation and diffusion pro-

cesses as the temperature approaches  $T_g$ , the glass-transition temperature [11–13].

These spatial heterogeneities occur over time scales that are long with respect to the basic unit time of the systems—one step in coupled maps and a lattice vibration in supercooled liquids—but shorter than the macroscopic relaxation, where materials are known to be homogeneous. As the temperature or the driving force is decreased, these regions become more and more stable and might overtake the whole sample at a transition temperature. Although the nature of spatial heterogeneities and their relation to the macroscopic dynamics remain incompletely understood, there is a considerable amount of work that relates directly to this issue. The mode-coupling theory describes the dynamics from the liquid phase into the supercooled regime and makes quantitative predictions that have been verified numerically in many systems [14,15]. It cannot yet describe accurately the very long time behavior in the supercooled regime, however [14]. Numerically, dynamical heterogeneities have been studied using a wide range of criteria tied to various aspects of this feature [5,16,17]. Recently, however, some groups have focused on the renewal or mean first-passage time [18] and the waiting-time distribution, which provide some link to the structure of the energy landscape [19,20].

Although both the dynamical heterogeneities and the stretched exponential behavior are characteristics of many frustrated systems (e.g., supercooled liquids [21]), it is not clear what the relation between these two properties is. In this paper, we address this question and show that dynamical heterogeneities can be directly responsible for the observation of stretched exponential relaxation in coupled map lattices. We also show that these results are applicable to supercooled liquids and can provide a microscopic basis for the stretched exponential relaxation in these systems, in line with the observations of Denny *et al.* on the long time dynamics of metabasin hopping [20].

Recently, Hunt *et al.* found that the distribution of the renewal time of a certain stochastic process measured in a one-dimensional lattice of coupled diode resonators could be fitted to a stretched exponential function over six orders of magnitude [10]. Simulations on a related model of nonlinear maps diffusively coupled via short range interactions show

that the fit can be extended to more than nine orders of magnitude, ruling out any other power law or simple combination of pure exponentials [10,22]. The quality of the fit in this system is sufficient to distinguish the region of the parameter space where the dynamics is described by a power-law distribution coupled with an exponential cutoff from the really stretched exponential distributions. (Reference [22] contains background information important for understanding the results of the present work and will be referred to as I in the following.)

These simulations I also demonstrated the importance of the effect of the system's size on its dynamics. While stretched exponentials are sufficiently robust and are observed even in some small samples, the quality of such fits is much poorer than in the big samples. This is in direct analogy with real experimental results and supports the observation that the quality of the samples is crucial for obtaining unambiguous stretched exponential behavior and meaningful values of the exponent  $\beta$  [1,3].

Using simulations on this and other models, as well as some analytical results, we show here that (1) the stochastic process studied in Ref. [10] and I can be identified with a renewal process [23], (2) a stretched exponential distribution of the renewal time implies a similar shape for the decay of the two-point autocorrelation function, and (3) renewal processes are present in a supercooled liquid and play a central role in its dynamics.

Granted the universal features of SER, readers unfamiliar with chaos theory and coupled maps may not see immediately the relevance of these mathematical tools to understanding the dynamical properties of supercooled liquids and glasses. Even the successful predictions of the trap (sink) model [1,24] appear to be unexpected. For instance, the survival probability of a random walker in the presence of randomly distributed static traps crosses over from exponential to SER only at longer times where very few ( $\sim 10^{-30}$  for 10% traps) of the walkers have survived [25]. In our examples of steady-state dynamics in coupled maps, the analogous crossover in trap-time distributions  $\rho(t)$  occurs near  $10^{-1}$  to  $10^{-2} \times \max\{\rho(t)\}$ , depending on the value of a control parameter. Even the latter number is small compared to the crossovers observed, for example, for relaxation of the first peak in the structure factor in molecular dynamics simulations (MDSs) of crystallization-avoiding soft sphere mixtures [26], which occur near 3/4 of the maximum value. The reason for these differences is that the systems chosen for MDSs already represent prepared states with strong glass-forming tendencies. The random walker with random traps model contains spatial inhomogeneities, which we will show represent an essential feature necessary for SER, and which are absent from some popular models of static glass structure such as mode-coupling theory [27]. That model does not contain the collapse of phase space that occurs as the liquid is supercooled to the glass transition, and that is why SER appears only at very long times. However, it does contain the correct dimensional dependence of diffusive behavior that leads to the relation  $\beta = d/(d+2)$ , which is important for comparisons to experiment. That the one-dimensional coupled map lattice discussed here reaches a steady state

much more rapidly than the random walker is a clear indication that this model is physically relevant. The way in which this happens was illustrated in Fig. 8 of I: the surviving periodic domains are those that have avoided chaotic regions.

We also emphasize that in this simple model the behavior of the stretching exponent  $\beta$  as a function of a control parameter  $r$  is similar to the dependence of  $\beta$  on the temperature in both three- (3D) [28] and one-dimensional (1D) [8] spin-glass models. This similarity captures not only the qualitative tendency for  $\beta$  to decrease with decreasing  $r$  (or  $T$ ), but also the quantitative agreement in the low- $r$  (or low- $T$ ) limit  $\beta \approx 1/3$ . The same lowest value of  $\beta$  was obtained in studies of random walks on fractals [29–31].

## II. RESULTS

### A. Properties of the coupled map model

The nonlinear model of Ref. [10], used to reproduce qualitatively the experimental system, is a one-dimensional chain of  $N$  diffusively coupled nonlinear deterministic maps,  $f(x)$ , with a coupling constant  $\alpha$  and periodic boundary conditions. The time evolution of this system is discrete and is described by the following iterative equation:

$$x_n(t+1) = (1-\alpha)f(x_n(t)) + \frac{\alpha}{2}\{f(x_{n-1}(t)) + f(x_{n+1}(t))\}, \quad (1)$$

which, given initial conditions  $x_n(0)$  for each site  $n = 1, 2, \dots, N$ , generates a time series  $\{x_n(t)\}$ . The interaction is totalistic and involves only the nearest neighbors. Following I, in this study we use the logistic map  $f(x) = rx(1-x)$ .

Following the analysis of the experimental data, the stochastic process of interest is represented by a coarse-grained variable defined as

$$\sigma_n(t) = \text{sgn}[x_n(t) - x_{\text{thr}}], \quad t = 0, 2, 4, \dots, \quad (2)$$

where the quantity  $x_n(t)$  is defined in Eq. (1) and  $x_{\text{thr}}$  is a certain threshold value. The basic dynamics of the coupled oscillators being period 2, the analysis is also done over even (or odd) time steps.

This model shows a stretched exponential distribution of the renewal (or trap) times, which are defined in Sec. II B, over a wide range of the control parameter  $r$ , which plays a role akin to temperature as can be seen in Fig. 1. This figure shows the stretching exponent  $\beta_0$  and the time scale  $\tau_0$  as functions of the control parameter  $r$ . The values of  $\beta_0$  and  $\tau_0$  were obtained by least-squares fitting of the long-time part of the trap-time distributions for a chain of  $N = 10\,000$  coupled logistic maps  $f(x) = rx(1-x)$  with the expression  $A \exp[-(t/\tau_0)^{\beta_0}]$ . Although  $\beta_0(r)$  and  $\tau_0(r)$  are nonmonotonic, the overall behavior is similar to the results in both 3D [28] and 1D [8] spin-glass models. This broad similarity captures not only the qualitative tendency for  $\beta$  to decrease with decreasing  $r$  (or  $T$ ), but also the quantitative agreement in the low- $r$  (or low- $T$ ) limit  $\beta \approx 1/3$ , the same limit as that obtained in studies of random walks on fractals [29–31].

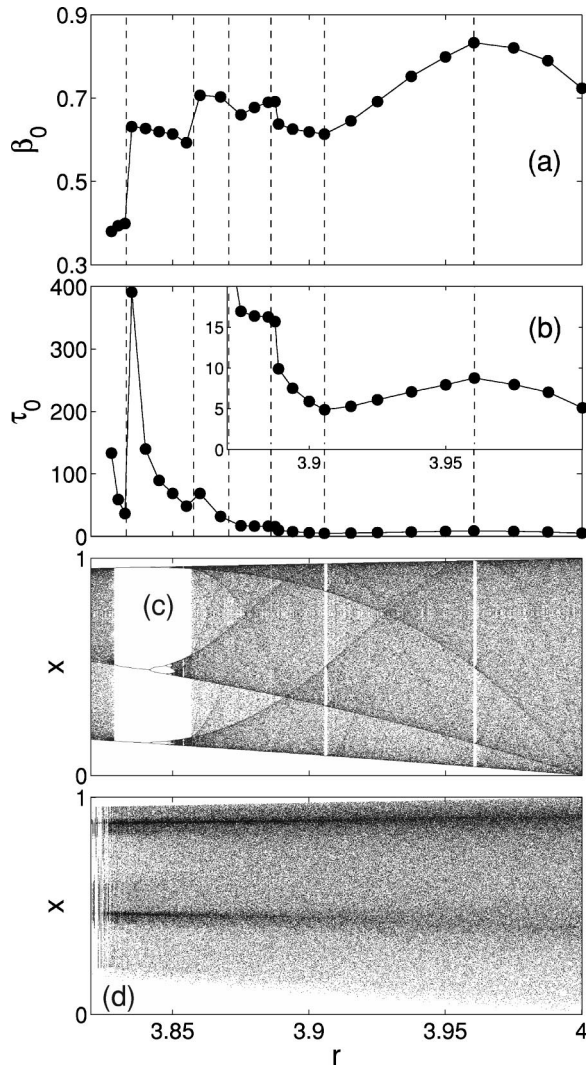


FIG. 1.  $\beta_0$  (a) and  $\tau_0$  (b) as functions of the logistic map parameter  $r$ . Dots correspond to the least-squares-fit values and the thin solid line is drawn as a guide to the eye. The vertical dashed lines demarcate the discontinuities in  $\beta_0(r)$  and  $\tau_0(r)$ . They correspond very well to the positions of the periodic cascades in the bifurcation diagram of the uncoupled logistic map shown in (c). (d) shows the bifurcation diagram for a site in the coupled map lattice. Inset in (b) magnifies the right hand side of the main plot where the variation of  $\tau_0$  with  $r$  is not distinguishable. The accuracy of  $\beta_0$  and  $\tau_0$  obtained by a least-squares fit to the long-time part of the trap-time distribution is within 15%.

Although the bifurcation diagram for a site in the coupled map lattice Fig. 1(d) appears to be uniform with respect to  $r$  down to  $r \approx 3.8275$ , the nonmonotonicity of  $\beta_0(r)$  and  $\tau_0(r)$  most probably arises from the fact that for the isolated logistic map and the values of  $r < 4$  chaotic orbits are interspersed with period cascades as seen in Fig. 1(c), i.e., the approach to full chaos as  $r$  goes to 4 is not uniform. (For an explanation of the concept of bifurcation diagrams, see, e.g., Ref. [32].) Interestingly, the lowest value of  $\beta_0$  is attained within the widest window of the period-3 cascade in the bifurcation diagram of the uncoupled logistic map. About and below  $r = 3.82$ , in the coupled map chain, very narrow chaotic win-

dows are also interspersed with periodic ones, and the stretched exponential behavior of the trap-time distribution, with  $\beta_0$  greater than or about  $1/3$ , is recovered only for some values of  $r$ . This interesting behavior suggests that SER is uncovering an additional aspect of the dynamics of coupled map lattices. A full analysis of this behavior lies outside the framework of this paper, but we plan to return to studying it elsewhere.

In other models and experiments, the time scale  $\tau$  grows monotonically with decreasing temperature. It is not the case here, although the values of  $\tau_0$  are greatest at the low- $r$  (and low- $\beta_0$ ) end, following the general trend observed elsewhere.

## B. Renewal processes in coupled maps

The stochastic process  $\sigma_n(t)$  is a renewal process [23] provided that the time intervals (renewal times) between two subsequent zero crossings are statistically independent random variables. We tested the independence of these time intervals for the present model, Eq. (2), by calculating the distributions of the time intervals following time intervals of a specified length. The distributions were identical regardless of the length of the preceding time intervals.

In general, a renewal process is defined so that  $\sigma(t) = +1$  for  $t_0 < t \leq t_1$ ,  $\sigma(t) = -1$  for  $t_1 < t \leq t_2$ ,  $\sigma(t) = +1$  for  $t_2 < t \leq t_3$ , and so on. The renewal processes are the simplest possible stochastic processes readily susceptible of probability theoretical analysis [33]. The dynamics represented by such processes can be understood in terms of transition probabilities between the two possible states  $\sigma = \pm 1$  [34].

In I, it is shown that the existence of very long renewal time intervals  $t_{n+1} - t_n$  associated with the stretched exponential distribution can be attributed to the presence of time-limited ordered domains (or traps) with a dominant spatial period of four sites. Figure 2(a) shows a typical distribution of the renewal time intervals (or trap times) for this model.

These results suggest that the dynamical heterogeneity manifested by the presence of time-limited ordered domains could be at the origin of the stretched exponential relaxation in supercooled liquids and glasses, and the renewal processes could provide a simple picture for describing heterogeneous dynamics in these condensed phases. In order to support these two affirmations, we first need to show that (1) there is a one-to-one correspondence between renewal processes and their time autocorrelation functions, because experimentally measurable relaxation responses are mathematically described by autocorrelation functions of certain dynamical variables and not directly by nonobservable renewal processes, and (2) the renewal processes are present in supercooled liquids and glasses and can be calculated effectively, exhibiting unambiguous stretched exponential distributions of renewal time intervals.

The relation between the autocorrelation function

$$C(t) \equiv \langle \sigma(t') \sigma(t'+t) \rangle_{t'}, \quad (3)$$

of a renewal process  $\sigma(t)$  and the distribution  $\rho(t)$  of time intervals  $t_{n+1} - t_n$  between the zero crossings (or, in other words, renewals) of this process was recently studied for a

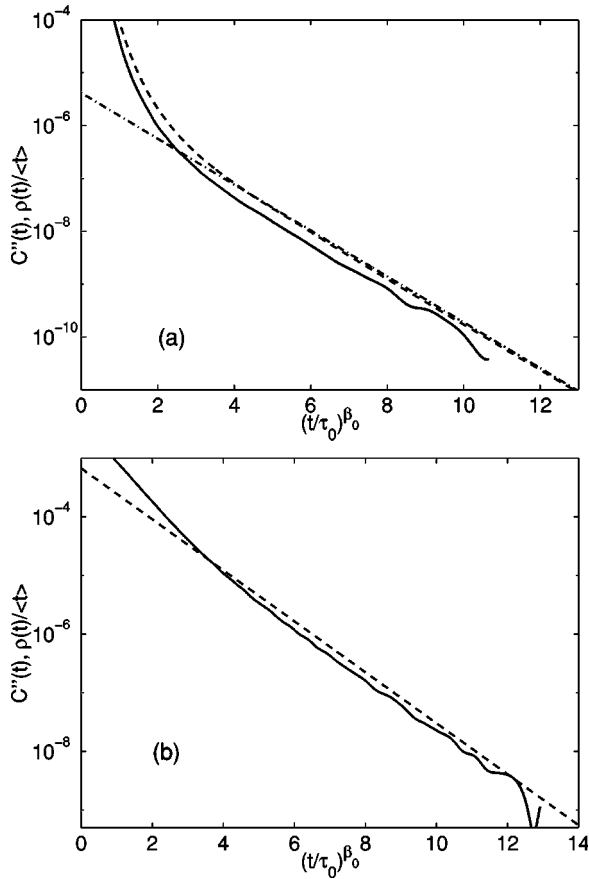


FIG. 2. (a) Second derivative  $C''(t)$  of the autocorrelation function  $C(t)$  of the coarse-grained orbit  $\sigma(t)$ , Eq. (2), with  $r=3.83$  (solid line) compared to the asymptotic behavior  $\rho(t)/\langle t \rangle$  given by Eq. (8) (dashed line). Dash-dotted line shows the stretched exponential fit to the long-time part of  $\rho(t)$  with  $\tau_0=59\pm 9$  and  $\beta_0=0.39\pm 0.05$ . (b) The same as in (a), but for the renewal process computer generated with the renewal times distributed according to Eq. (4) with  $\tau_0=5$  and  $\beta_0=0.4$ . In this case, the lines corresponding to the dashed and dash-dotted lines in (a) coincide.

range of distributions [23]. Following this approach, Godrèche and Luck also showed [35] that  $C(t)$  corresponding to the stretched exponential distribution

$$\rho(t) = \frac{\beta_0}{\tau_0 \Gamma(\beta_0^{-1})} \exp[-(t/\tau_0)^{\beta_0}] \quad (4)$$

can also be represented by a stretched exponential at long times:

$$C(t) \approx C_{SE}(t) \equiv A \exp[-(t/\tau)^\beta], \quad t \rightarrow \infty. \quad (5)$$

We can Laplace transform the distribution of intervals  $\rho(t)$ , obtaining a function with a small singular part,  $\hat{\rho}(u) = 1 - \langle t \rangle u + \dots + \hat{\rho}_{\text{sing}}(u)$ ,  $u \rightarrow 0$  [36], where  $\langle t \rangle = \tau_0 \Gamma(2/\beta_0) / \Gamma(1/\beta_0)$  is the first moment of  $\rho(t)$ . From Ref. [23], we also know that the Laplace transform of the autocorrelation function is given by

$$\hat{C}_{\text{eq}}(u) = \frac{1}{u} \left( 1 - \frac{2[1 - \hat{\rho}(u)]}{\langle t \rangle u [1 + \hat{\rho}(u)]} \right). \quad (6)$$

Using the expression above for  $\hat{\rho}(u)$ , one finds, for the singular part of the correlation function at equilibrium,

$$\hat{C}_{\text{eq,sing}}(u) \approx \frac{\hat{\rho}_{\text{sing}}(u)}{\langle t \rangle u^2}. \quad (7)$$

Since the behavior at large times is governed by its singular part, we get

$$C''_{\text{eq}}(t) \approx \rho(t) / \langle t \rangle. \quad (8)$$

This implies that for a stretched exponential  $\rho(t)$  in the asymptotic limit  $C_{\text{eq}}(t) \propto \rho(t)$  [35].

The range of relevance for this analytical result can be confirmed numerically by studying the statistical properties of a computer-generated sequence of (pseudo)random numbers distribution according to the stretched exponential probability density function (PDF) Eq. (4). Using the fundamental transformation law of probabilities [37], given a computer-generated sequence of (pseudo)random numbers  $\{\theta\}$  distributed uniformly in the interval  $0 \leq \theta \leq 1$ , we apply a transformation rule  $t = \tau_0 [Q^{-1}(1/\beta_0, \theta)]^{1/\beta_0}$  so that the resulting sequence  $\{t\}$  is distributed according to the stretched exponential PDF  $\rho(t)$ . In order to compute the inverse regularized incomplete gamma function  $Q^{-1}(a, \theta)$ , we used the software from Refs. [38,39].

Given a sequence of time intervals distributed according to a prescribed PDF, it is straightforward to construct the corresponding renewal process and its autocorrelation function and second derivative. The second derivative is calculated numerically, after applying a smoothing function before each derivative to improve the quality of the operation.

The resulting  $C''(t)$  and  $C(t)$ , shown in Figs. 2 and 3, respectively, demonstrate clearly that the correspondence between  $\rho(t)$  and  $C''(t)$  holds for long times for  $C(t)$  calculated both for the coupled map lattice model, Eq. (1), with  $f(x) = rx(1-x)$ ,  $r=3.83$ ,  $\alpha=0.25$ , and  $N=1000$ , and for a renewal process where the renewal time-interval distribution is stretched exponential by prescription.  $C''(t)$  obtained numerically agrees with  $\rho(t)/\langle t \rangle$ , Eq. (8), without any free parameter within a multiplicative factor of the order of unity.

Interestingly, for the range measured,  $C(t)$  also follows the stretched exponential form, albeit with somewhat different parameters  $\tau_c$  and  $\beta_c$ . This indicates that the true asymptotic regime for  $C(t)$  or  $\rho(t)$  may be reached at longer time with a gradual approach toward this long-time behavior. This agrees with the theories (see, e.g., [24]) where the stretched exponential functions are accompanied by time-dependent multiplicative factors. For practical purposes, however, it is remarkable that the stretched exponential fit to the correlation function agrees with  $C(t)$ , starting with a value well observed experimentally between  $3/4$  and  $1/2 \times \max\{C(t)\}$ , and following with very high accuracy for several orders of magnitude to the regime beyond the reach of the experiments.

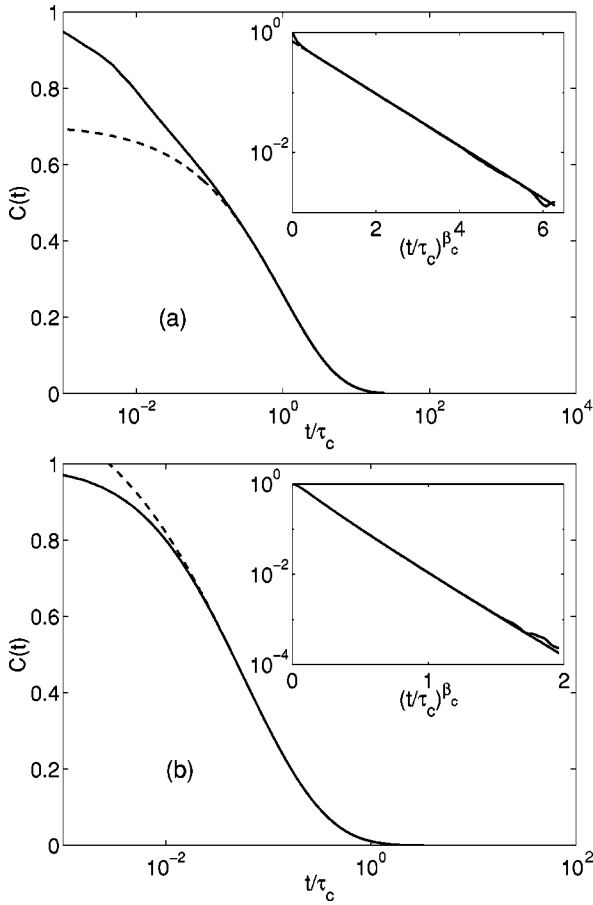


FIG. 3. (a) Autocorrelation function  $C(t)$  of the coarse-grained orbit  $\sigma(t)$ , Eq. (2), with  $r=3.83$  (solid line) and the stretched exponential fit to the long-time part of  $C(t)$  with  $\tau_c \approx 1250$  and  $\beta_c \approx 0.58$  (dashed line). The inset shows the same curves, but with a logarithmic vertical axis. (b) The same as in (a), but for the renewal process computer generated with the renewal times distributed according to Eq. (4) with  $\tau_0=5$  and  $\beta_0=0.4$ . In this case the stretched exponential fit to  $C(t)$  is with  $\tau_c \approx 69$  and  $\beta_c \approx 0.54$ .

### C. Molecular dynamics

Having established the correspondence between the distribution of time intervals of renewal processes and their autocorrelation functions, we now show that a renewal process can also be identified in a supercooled liquid, and it appears to provide a simple and elegant description for the stretched exponential relaxation. In order to do so, we have performed molecular dynamics simulations of a 16 000-particle single-component system where the interactions between the particles are described by a pair potential labeled Z2 in Ref. [40]. This potential is similar to that introduced initially by Dzугutov [41] but provides a longer-lived metastable supercooled state prior to crystallization. Otherwise the properties of the Z2 liquid are similar to those of the Dzугutov liquid; in particular, it exhibits dynamical heterogeneities formed by short-lived clusters composed of connected icosahedra [42]. The simulations are performed at  $T=0.68$ , below the melting temperature of  $T_m=0.70 \pm 0.05$ , with a time step of 0.01 in reduced Lennard-Jones units [43]. We take measurements over 16 000 atoms with the number density  $\rho=0.85$  for 7

$\times 10^6$  time steps, stopping before the system starts crystallizing. This simulation time would correspond to a 150 ns simulation for argon.

A renewal process representing the atomic dynamics in the liquid can be constructed so that each renewal time interval corresponds to the time a particle spends in a dynamical trap. This is identical to the first-passage time, in the parlance of Allegrini *et al.* [18], but differs formally from the waiting time of Doliwa and Heuer [19] and Denny *et al.* [20], which focuses on hops between energy basins. We say that particle  $i$  is trapped as long as its displacement

$$d_i(t) = |\mathbf{r}_i(t) - \mathbf{r}_i(0)| \quad (9)$$

measured starting from an arbitrary initial position  $\mathbf{r}_i(0)$  remains less than a certain threshold value  $d_{\text{thr}}$ . The moment  $t=t_{\text{thr}}$  of surpassing the threshold is counted as a renewal and the value of  $\mathbf{r}_i(0)$  is reset to  $\mathbf{r}_i(t_{\text{thr}})$  as well as the time origin for the next trap.

The corresponding trap-time distribution (see Fig. 4) is not particularly sensitive to the choice of  $d_{\text{thr}}$  as long as it is of the order of the effective atomic diameter. To be specific, we choose  $d_{\text{thr}} = 2\pi/Q_0 \approx 0.88$  [43], where  $Q_0$  is the position of the main peak in the static structure factor  $S(Q)$  and neglect the temperature dependence of this length scale. A threshold of  $\approx 0.88$  is about four times larger than the critical value estimated by Allegrini *et al.* in a binary Lennard-Jones system. While the focus of Ref. [18] is on the inertial regime, which takes place at small distances, on the order of 0.1 atomic radius, we are interested in the atomic displacement leading to a change in the configuration. Figure 4(a) also shows in the inset that the trap-time distribution is exponential at a temperature well above the melting point ( $T_m = 0.70 \pm 0.05$ ) and its long-time tail well agrees with a stretched exponential fit with  $\beta_0 \approx 0.71$  in the supercooled regime. In order to be considered as a fully Markovian process, we also assess the correlation between subsequent trapping time intervals, as was done for the coupled map lattice. Comparing trap distributions considering only the subset of events following traps of various length, again, we find no correlation between renewal time intervals. The value of  $\beta_0$  agrees well with  $\beta = 0.70 \pm 0.05$  for the self part of the intermediate scattering function  $F_s(Q_0, t)$  calculated at the same temperature and for the same length scale. [For the definition of  $F_s(Q, t)$  see, e.g., Ref. [44].]  $F_s(Q_0, t)$  for the Z2 liquid behaves similarly to the same quantity calculated for the supercooled Dzугutov liquid [45] and is not shown here.

These results appear at odds with the analysis of Allegrini *et al.*, who report a power-law distribution for the long-time tail of the renewal time [18], although admitting that the asymptotic cutoff in the distributions is difficult to resolve numerically. Figure 4(b) shows our simulation results at  $T=0.68$  plotted on a log-log scale with a tentative fit of the long-time behavior. It is clear that the stretched exponential provides a much better fit to the simulation data. This difference between the two simulations might be due to both the physical range of parameters and the precision of the simulation. First, Allegrini and collaborators focus on the inertial length scale while the results presented here require atomic

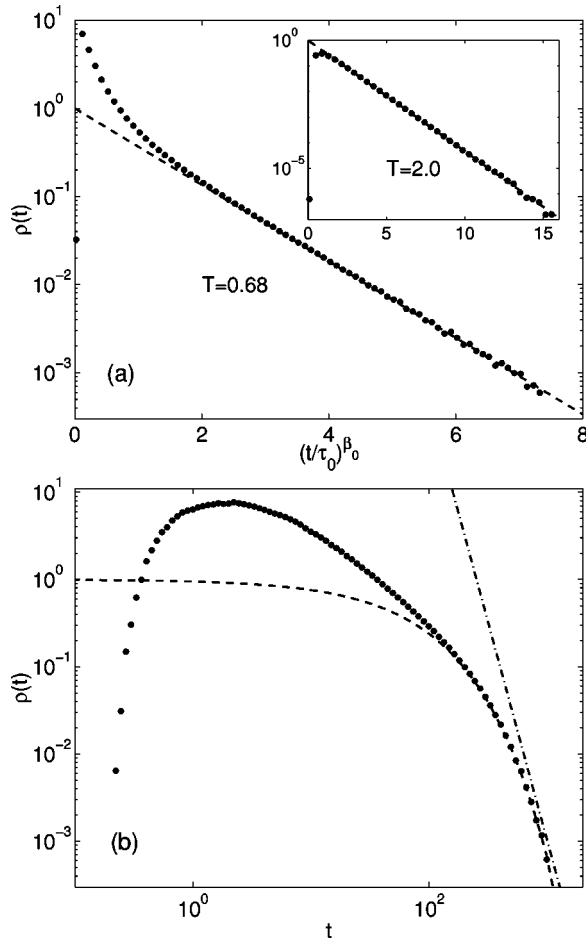


FIG. 4. (a) Dots: distributions of trap-time intervals for low ( $T = 0.68$ , each data point corresponds to an average over 500 time steps) and high temperatures (inset,  $T = 2$ ). Dashed lines: corresponding stretched exponential fits with  $\tau_0 = 60 \pm 5$ ,  $\beta_0 = 0.71 \pm 0.05$  and (inset)  $\tau_0 = 1.22 \pm 0.05$ ,  $\beta_0 = 1 \pm 0.002$ . (b) Dots: the same as in (a) for  $T = 0.68$  presented on a log-log scale over a mesh with logarithmic time intervals. Dashed line: the same as in (a) for  $T = 0.68$ . Dash-dotted line: power law  $\propto 1/t^5$ .

diffusion, not only thermal vibrations; second, focusing on a single temperature, our simulation uses a larger cell and averages over a time scale roughly six times longer.

Denny *et al.* [20] also note that the distribution in Ref. [18] may not be a power law. Although the waiting time distribution in Ref. [20] differs formally from that studied here, at long times they can be comparable since the escape of a particle from a sphere of radius of the order of the atomic diameter is likely to correspond to the transition of the phase point in the configuration space between energy metabasins. We note, however, that the log-normal form of the waiting time distribution proposed by Denny *et al.* results in a correlation function that can only be *approximated* with SER, whereas the stretched exponential first-passage-time distribution *results* in a correlation function that is also a stretched exponential at long time.

### III. CONCLUSION AND DISCUSSION

As was shown in I, traps in coupled map lattices correspond to ordered short-lived domains. In direct analogy, it is

natural to expect that the trap condition  $d_i(t) < d_{\text{thr}}$ , for  $d_i(t)$  from Eq. (9), is satisfied as long as particle  $i$  is literally trapped within a relatively stable environment—a “cage.” The renewal process formed in this way and the associated trap-time distribution and autocorrelation function provide a simpler description of the structural relaxation in a condensed matter system than, e.g., the cage correlation function [46] recently used to demonstrate the stretched exponential relaxation in a Lennard-Jones massively defective crystal.

In particular, traps in the coupled map lattice show only a very short spatial extension that does not seem to diverge as the overall dynamics of the system slows down. As  $\beta_0$  goes from  $0.6 > \beta_0 > 0.5$  to  $0.4 > \beta_0 > 0.3$ , the longer traps go from about 120 to more than 30 000 time steps but the characteristic time-averaged width of these traps only quadruples, from two to eight sites (see I). These observations are consistent with the behavior of dynamical heterogeneities observed in a number of model systems [7,8,16,42].

We note that the only model that rigorously derives the stretched exponential long-time asymptotic behavior is that of a random walker in a system with static traps (see, e.g., Ref. [1]). Reference [24] cites both upper [47] and lower [48] bounds for the asymptotic behavior of the survival probability. Both bounds have the same  $\beta$ , but different  $\tau$ . It is not obvious, however, that the above theory directly applies to the results of this work. Nevertheless, our results are obtained in terms of first-passage statistical distributions of the type encountered in the static-trap model, and the use of similar mathematical techniques may shed more light on the nature of the dynamics in the models presented here.

In conclusion, we focused on the relation between dynamical heterogeneities and stretched exponential relaxation in two models: a coupled chaotic oscillator system and a supercooled liquid. Without insisting that the former model represents the actual behavior of real liquids, we have shown that the asymptotic long-time behavior of the autocorrelation function of a renewal process is a stretched exponential provided that the distribution of the renewal time intervals  $\rho(t)$  is also a stretched exponential at long times, i.e.,  $\rho(t) \approx \exp[-(t/\tau_0)^{\beta_0}]$ . Using this relation, we have demonstrated that, at least in the two model systems considered here, the relaxation dynamics can be described in terms of well-defined dynamical traps, providing useful insight regarding the absence of a diverging static length scale at the glass transition.

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- [1] J.C. Phillips, Rep. Prog. Phys. **59**, 1133 (1996).
- [2] J.C. Phillips, cond-mat/0304501e-print .
- [3] J.C. Phillips, Physica C **340**, 292 (2000).
- [4] J.C. Phillips and J.M. Vandenberg, J. Phys.: Condens. Matter **9**, L251 (1997).
- [5] C. Donati, S.C. Glotzer, P.H. Poole, W. Kob, and S.J. Plimpton, Phys. Rev. E **60**, 3107 (1999).
- [6] P.G. Debenedetti and F.H. Stillinger, Nature (London) **410**, 259 (2001).
- [7] J.P. Garrahan and D. Chandler, Phys. Rev. Lett. **89**, 035704 (2002).
- [8] M.L. Mansfield, Phys. Rev. E **66**, 016101 (2002).
- [9] A.M.S. Bhattacharyya and B. Bagchi, J. Chem. Phys. **117**, 2741 (2002).
- [10] E. Hunt, P. Gade, and N. Mousseau, Europhys. Lett. **60**, 827 (2002).
- [11] R. Richert, J. Phys.: Condens. Matter **14**, R703 (2002).
- [12] H. Sillescu, J. Non-Cryst. Solids **243**, 81 (1999).
- [13] A. Cristanti and F. Ritort, Philos. Mag. B **82**, 143 (2002).
- [14] S.-H. Chong, W. Götze, and M.R. Mayr, Phys. Rev. E **64**, 011503 (2001).
- [15] S.-H. Chong and W. Götze, Phys. Rev. E **65**, 051201 (2002).
- [16] K. Vollmay-Lee, W. Kob, K. Binder, and A. Zippelius, J. Chem. Phys. **116**, 5158 (2002).
- [17] D. Caprion, J. Matsui, and H.R. Schober, Phys. Rev. Lett. **85**, 4293 (2000).
- [18] P. Allegrini, J.F. Douglas, and C. Glotzer, Phys. Rev. E **60**, 5714 (1999).
- [19] B. Doliwa and A. Heuer, Phys. Rev. E **67**, 030501 (2003).
- [20] R.A. Denny, D.R. Reichman, and J.P. Bouchaud, Phys. Rev. Lett. **90**, 025503 (2003).
- [21] X.Y. Xia and P.G. Wolynes, Phys. Rev. Lett. **86**, 5526 (2001).
- [22] S. Simdyankin, N. Mousseau, and E.R. Hant, Phys. Rev. E **66**, 066205 (2002).
- [23] C. Godrèche and J.M. Luck, J. Stat. Phys. **104**, 489 (2001).
- [24] B.D. Hughes, *Random Walks and Random Environments* (Clarendon Press, Oxford, 1995), Vol. 1.
- [25] G. Barkema, P. Biswas, and H. van Beijeren, Phys. Rev. Lett. **87**, 170601 (2001).
- [26] J.N. Roux, J.L. Barrat, and J.-P. Hansen, J. Phys.: Condens. Matter **1**, 7171 (1989).
- [27] W. Götze and L. Sjögren, Rep. Prog. Phys. **55**, 241 (1992).
- [28] A.T. Ogielski, Phys. Rev. B **32**, 7384 (1985).
- [29] I. Campbell, J. Phys. (France) Lett. **46**, L1159 (1985).
- [30] R. de Almeida, N. Lemke, and I. Campbell, Eur. Phys. J. B **18**, 513 (2000).
- [31] P. Jund, R. Jullien, and I. Campbell, Phys. Rev. E **63**, 036131 (2001).
- [32] E. Ott, *Chaos in Dynamical Systems* (Cambridge University Press, Cambridge, England, 1993).
- [33] W. Feller, *An Introduction to Probability Theory and its Applications*, 3rd ed. (Wiley, New York, 1970), Vol. 1.
- [34] G. Gielis and C. Maes, Europhys. Lett. **31**, 1 (1995).
- [35] C. Godrèche and J.M. Luck (private communication).
- [36] The following analysis also holds if  $\rho(t)$  is a Lévy law.
- [37] L. Devroye, *Non-Uniform Random Variate Generation* (Springer-Verlag, New York, 1986).
- [38] A. DiDonato and J.A.H. Morris, ACM Trans. Math. Softw. **13**, 318 (1987); URL <http://doi.acm.org/10.1145/29380.214348>
- [39] A. DiDonato and J.A.H. Morris, ACM Trans. Math. Softw. **12**, 377 (1986).
- [40] J.P.K. Doye, D.J. Wales, F.H.M. Zetterling, and M. Dzugutov, J. Chem. Phys. **118**, 2792 (2003).
- [41] M. Dzugutov, Phys. Rev. A **46**, R2984 (1992).
- [42] M. Dzugutov, S.I. Simdyankin, and F.H.M. Zetterling, Phys. Rev. Lett. **89**, 195701 (2002).
- [43] Following Refs. [40,41], we use reduced Lennard-Jones units [49] for all quantities here.
- [44] J.-P. Hansen and I.R. McDonald, *Theory of Simple Liquids*, 2nd ed. (Academic Press, London, 1986).
- [45] M. Dzugutov, Europhys. Lett. **26**, 533 (1994).
- [46] E. Rabani, J.D. Gezelter, and B. Berne, Phys. Rev. Lett. **82**, 3649 (1999).
- [47] R.F. Kayser and J.B. Hubbard, Phys. Rev. Lett. **51**, 79 (1983).
- [48] P. Grassberger and I. Procaccia, J. Chem. Phys. **77**, 6281 (1982).
- [49] M.P. Allen and D.J. Tildesley, *Computer Simulation of Liquids* (Clarendon Press, Oxford, 1987).