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The RARE model: A generalized approach to random relaxation processes in disordered systems

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This paper introduces and analyses a general statistical model, termed the RAndom RElaxations (RARE) model, of random relaxation processes in disordered systems. The model considers excitations that are randomly scattered around a reaction center in a general embedding space. The model's input quantities are the spatial scattering statistics of the excitations around the reaction center, and the chemical reaction rates between the excitations and the reaction center as a function of their mutual distance. The framework of the RARE model is versatile and a detailed stochastic analysis of the random relaxation processes is established. Analytic results regarding the duration and the range of the random relaxation processes, as well as the model's thermodynamic limit, are obtained in closed form. In particular, the case of power-law inputs, which turn out to yield stretched exponential relaxation patterns and asymptotically Paretian relaxation ranges, is addressed in detail. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4770266]

I. INTRODUCTION

Relaxation, the return of a perturbed system into equilibrium, is one of the most fundamental processes of physical systems. The simplest relaxation model is the exponential or Debye law $\exp(-t/\tau)$ incorporating the relaxation time scale τ. In many cases, however, significant deviations from the exponential law have been observed. The two most prominent generalized relaxation laws to accommodate these deviations are the stretched exponential law $\exp(-[t/\tau]^{\alpha})$ with $0 < \alpha < 1$ and the asymptotic power or Pareto law (sometimes also called Nutting law) $(1 + [t/\tau])^{(-\beta)}$ with $\beta > 0$. When compared to the simple exponential law, both these generalized laws correspond to a broad distribution of relaxation times.[1] Generalized relaxation patterns are observed on many scales, ranging from single molecules[2–4] over dielectric response[5] to macroscopic viscoelasticity.[6–9]

Theoretical approaches to describe generalized relaxation processes include parallel relaxation channels,[10,11] hierarchically constrained dynamics giving rise to complex serial relaxation,[12] and defect diffusion models.[13] For the stretched exponential law Klafter and Shlesinger demonstrated the common universal features behind these approaches.[14] There exist also extensions of stretched exponentials in models of dynamic relaxation channels.[15] Yet another approach to generalized relaxation dynamics is that of fractional-order, viscoelastic mechanical bodies;[16–20] for their historical development see Ref. 21. Similar methods based on generalized dynamic equations and stochastic approaches based on stable distributions have been used to describe dielectric relaxation behavior.[22,23]

In this paper, we establish a robust Poissonian approach to complex relaxation processes based on individual reactions between excitations that are randomly scattered around a reaction center in a general embedding space. The resulting model is termed RARE, the acronym standing for RAndom RElaxations. The model's input quantities are the spatial scattering statistics of the excitations around the reaction center, and the chemical reaction rates between the excitations and the reaction center as a function of their mutual distance. The RARE model is considerably general, it has a versatile framework, and it allows for a fairly intuitive interpretation of complex relaxation processes. The RARE model is analyzed in detail, and analytic results regarding the duration and the range of the model's random relaxation processes are obtained in closed form.

The paper is organized as follows. The RARE model is introduced, intuitively explained, and rigorously constructed in Sec. II. The detailed stochastic analysis of the RARE model and a Monte-Carlo algorithm for the simulation of the model's random relaxation processes are presented, respectively, in Secs. III and IV. The results presented in Sec. III establish a comprehensive statistical picture of the duration and the range of the model's random relaxation processes: marginal distributions, joint distribution, and conditional distributions. The case of power-law inputs, which are shown to yield stretched exponential relaxation patterns and asymptotically Paretian relaxation ranges, is addressed in Sec. V. The thermodynamic limit of the RARE model is investigated in Sec. VI. Detailed proofs of the results stated along the paper are given in the Appendixes.

II. THE RARE MODEL

The RARE model we introduce and explore in this paper is described as follows. A reaction center is placed at an arbitrary point P of a general metric space $\mathcal{M}$, and a countable collection of excitations is scattered randomly across the

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space $\mathcal{M}$. The excitations are labeled with the index i, and the position of excitation i is the random point P_i. The distance in the metric space $\mathcal{M}$ is measured by a general metric function $\mathbf{d}(\cdot,\cdot)$, and the distance between the reaction center and excitation i is $D_i = \mathbf{d}(P_i, P)$. Excitation i is equipped with a random timer T_i, and a reaction between the center and the excitations occurs upon the first timer-expiration event. The model is statistically characterized by two random variables, the *reaction time* T and the *reaction range* X, which are defined as follows: The *reaction time* T is the time elapsing until the first timer expires

$$T = \min_i\{T_i\}. \tag{1}$$

The *reaction range* X is the distance between the reaction center and the excitation whose timer first expired

$$X = \sum_i D_i \mathbf{I}(T = T_i). \tag{2}$$

In Eq. (2) and hereinafter, $I(E)$ denotes the indicator function of an event E (i.e., $I(E) = 1$ if the event did occur, and $I(E) = 0$ if the event did not occur).

Setting the space $\mathcal{M}$ in which the reaction takes place to be a metric space yields great versatility. Indeed, $\mathcal{M}$ can be an Euclidean space of arbitrary dimension, a non-Euclidean space, such as an elliptic space or a hyperbolic space, a general surface or landscape, a fractal object, a network, etc. We now turn to specify the RARE model assumptions, and thereafter present a preliminary analysis.

A. Model assumptions

To quantify the RARE model the distributions of the random points $\{P_i\}$, as well as the distribution of the random timers $\{T_i\}$, need to be specified. A highly applicable and effective statistical methodology to model the random scattering of points, in general, spaces are *Poisson processes*.[24] Poisson processes have a wide span of applications ranging from insurance and finance[25] to queuing systems,[26] and from fractals[27] to power-laws.[30] We henceforth assume that the random points $\{P_i\}$ form a general Poisson process defined on the metric space $\mathcal{M}$. Consequently, the "displacement theorem" of the theory of Poisson processes (see Sec. 5.5 in Ref. 24) implies that the distances $\{D_i\}$ form a general Poisson process defined on the positive half-line $(0, \infty)$. In what follows we denote by $\rho(x)$ the average number of excitations, which are within a distance x of the reaction center,

$$\rho(x) = \mathbf{E}\left[\sum_i \mathbf{I}(D_i \le x)\right], \quad x \ge 0. \tag{3}$$

In Eq. (3) and in the following, $\mathbf{E}$ denotes the mathematical expectation. Namely, if ξ is a real-valued random variable governed by the probability density function $f_\xi(x)$ (x real), and $\phi(x)$ is a real-valued function defined on the real line, then $\mathbf{E}[\phi(\xi)] = \int_{-\infty}^{\infty} \phi(x) f_\xi(x) dx$. Moreover, in Eq. (3) the function $\rho(x)$ is monotone increasing and is assumed to start at zero: $\rho(0) = 0$. We note that the derivative $\rho'(x)$ is the *Poissonian intensity* of the Poisson process $\{D_i\}$.[24]

As noted in the Introduction, the most common statistical law for reactions in the physical sciences is the exponential law. Given the distances $\{D_i\}$ we now assume that the timers $\{T_i\}$ are independent and exponentially distributed random variables, and that the exponential distribution of timer T_i is determined by the distance D_i. In what follows we denote by $\eta(x)$ the exponential rate of the timers as a function of the distance variable x. Namely, given the distance D_i, the timer T_i is exponentially distributed with tail distribution function

$$\Pr(T_i > t | D_i) = \exp\{-\eta(D_i)t\}, \quad t \ge 0. \tag{4}$$

In other words, given the distance D_i, the timer T_i is exponentially distributed with mean $\mathbf{E}[T_i|D_i] = 1/\eta(D_i)$. Typically, the function $\eta(x)$ is monotone decreasing in the distance variable x.

The RARE model is quantified by the pair of functions introduced above, the *scattering function* $\rho(x)$ and the *reactivity function* $\eta(x)$. The scattering function $\rho(x)$ quantifies the underlying spatial scattering of the excitations, and the reactivity function $\eta(x)$ quantifies the underlying distance-dependent reaction rate. The "inputs" of the RARE model are the scattering function $\rho(x)$ and the reactivity function $\eta(x)$, and the "outputs" of the RARE model are the reaction time T and the reaction range X. In what follows we establish the statistics of the random outputs based on the given deterministic inputs.

The RARE model introduced herein can be viewed as a descendant of a unified donor-acceptor energy transfer model presented by Blumen[28] and further studied by Burlatsky and co-workers.[53] In its basic form this model considers an N-site lattice, in which each site is occupied by an excitation with probability p (independent of all other sites), and the reactions between the excitations and the reaction center are governed by Eq. (4). Thomas and co-workers[29] consider a model similar to the one developed herein, but with a uniform distribution of excitations. The RARE model generalizes these models, as it (i) implements the notion of Poisson processes, (ii) regards both the reaction time T and the reaction range X, and (iii) considers a non-uniform distribution of excitations. On the one hand, the implementation of a Poisson scattering of excitations in a general metric space allows for a high variability and versatility on the other hand, this Poisson modeling is highly tractable, as it quantifies all the spatial and scattering details (both potentially having infinitely many degrees of freedom) into one single function, the scattering function $\rho(x)$. Moreover, in this paper we provide a stochastic analysis of the reaction pair (T, X), whereas in Ref. 28 only the reaction time T was analyzed.

B. Preliminary analysis

In Sec. III, we present a detailed stochastic analysis of the RARE model. To facilitate the stochastic analysis of Sec. III, we present here a preliminary analysis. In what follows we set $R(x)$ to denote the aggregate exponential rate corresponding to excitations, whose distance from the reaction center is greater than x,

$$R(x) = \sum_i \eta(D_i)\mathbf{I}(D_i > x), \quad x \ge 0. \tag{5}$$

Note that the aggregate rate $R(x)$ is a stochastic process parameterized by the distance variable x.

Conditioned on the realizations of distances $\{D_i\}$, the aggregate rate $R(x)$ yields compact formulas for the conditional distributions of the reaction time T and the reaction range X. Indeed, the assumptions of the RARE model, combined with the statistical properties of the minima of independent exponential random variables,[31] implies that: (i) The conditional distribution of the reaction time T is given by the tail distribution function

$$\Pr(T > t|\{D_i\}) = \exp\{-tR(0)\}, \quad t \geq 0. \tag{6}$$

(ii) The conditional distribution of the reaction range X is given by the tail distribution function

$$\Pr(X > x|\{D_i\}) = \frac{R(x)}{R(0)}, \quad x \geq 0. \tag{7}$$

(iii) The joint conditional distribution of the reaction pair (T, X) is given by the joint tail distribution function

$$\Pr(T > t, X > x|\{D_i\}) = \exp\{-tR(0)\}\frac{R(x)}{R(0)}, \quad t, x \geq 0. \tag{8}$$

Note that given the realizations of the distances $\{D_i\}$ the reaction time T and the reaction range X turn out to be *independent* random variables. Indeed, the joint tail distribution function of the reaction pair (T, X) equals the product of the tail distribution functions of the reaction time T and the reaction range X,

$$\Pr(T > t, X > x|\{D_i\}) = \Pr(T > t|\{D_i\})\Pr(X > x|\{D_i\}), \quad t, x \geq 0. \tag{9}$$

III. STOCHASTIC ANALYSIS

The preliminary analysis presented in Sec. II provides us with the conditional distribution of the reaction pair (T, X), conditioned on the realizations of distances $\{D_i\}$. A stochastic analysis detailed in Appendixes A–D shifts us from the aforementioned conditional distribution to the (unconditional) distribution of the reaction pair (T, X). In this section, we present the key results of this stochastic analysis.

A. Reaction time

The tail distribution function of the reaction time T is given by

$$\Pr(T > t) = \exp\left(-\int_0^\infty [1 - \exp\{-\eta(x)t\}]\rho(dx)\right), \tag{10}$$

for $t \geq 0$. In turn, taking the limit $t \to \infty$ in Eq. (10) yields the probability that a reaction never takes place

$$\Pr(T = \infty) = \exp\{-\rho(\infty)\}. \tag{11}$$

Equation (11) implies that a reaction takes place with certainty, i.e., $\Pr(T < \infty) = 1$, if and only if the scattering function $\rho(x)$ diverges, which in turn occurs if and only if there are infinitely many excitations. Moreover, differentiating Eq. (10) with respect to the time variable t yields the probability density function of the reaction time T,

$$f_T(t) = \Pr(T > t)\int_0^\infty \exp\{-\eta(x)t\}\eta(x)\rho(dx), \quad t > 0. \tag{12}$$

It is straightforward to deduce from Eq. (12) that the probability density function $f_T(t)$ is monotone decreasing from the level $f_T(0) = \int_0^\infty \eta(x)\rho(dx)$ (which can be either finite or infinite) to the level $f_T(\infty) = 0$.

B. Temporal hazard rate

The *hazard rate* of reaction time T is defined in terms of

$$h_T(t) = \lim_{\delta \to 0}\frac{1}{\delta}\Pr(T \leq t + \delta|T > t) = \frac{f_T(t)}{\Pr(T > t)} \tag{13}$$

with $t > 0$. Namely, the hazard rate $h_T(t)$ is the realization rate of the random variable T at time t, provided that it has not already occurred up to time t. The hazard rate is most commonly used in applied probability and in reliability theory.[32–34] Substituting the tail distribution function $\Pr(T > t)$ of Eq. (10) and the probability density function $f_T(t)$ of Eq. (12) into Eq. (13) yields the hazard rate of the reaction time T,

$$h_T(\tau) = \int_0^\infty \exp\{-\eta(x)t\}\eta(x)\rho(dx), \quad t > 0. \tag{14}$$

It is evident from Eq. (14) that the hazard rate $h_T(t)$, analogous to the probability density function $f_T(t)$, is monotone decreasing from the level $h_T(0) = \int_0^\infty \eta(x)\rho(dx)$ (which can be either finite or infinite) to the level $h_T(\infty) = 0$. Note that a monotone decreasing hazard rate $h_T(t)$ implies that the realization rate of the reaction time T *diminishes* in the course of time. Namely, the longer we are waiting for the reaction time T to realize, the *lower* the likelihood that it occurs immediately.

C. Reaction range

The probability density function of the reaction range X is given by

$$f_X(x) = (1 - \mathbf{E}[\exp\{-\eta(x)T\}])\rho'(x), \quad x > 0. \tag{15}$$

Equation (15) implies that if the Poissonian intensity $\rho'(x)$ is a monotone decreasing function, then so is the probability density function $f_X(x)$. In general, however, the probability density function $f_X(x)$ itself is not necessarily monotone decreasing. Eq. (15) further implies that the tail distribution function of the reaction range X is given by

$$\Pr(X > x) = \int_x^\infty (1 - \mathbf{E}[\exp\{-\eta(y)T\}])\rho(dy), \quad x \geq 0. \tag{16}$$

Note that both the probability density function $f_X(x)$ and the tail distribution function $\Pr(X > x)$ of the reaction range X involve the Laplace transform of the reaction time T.

D. Reaction pair

The joint probability density function of the reaction pair (T, X) is given by

$$f_{(T,X)}(t,x) = \Pr(T > t)\exp\{-t\eta(x)\}\eta(x)\rho'(x), \quad t,x > 0. \tag{17}$$

Comparing the joint probability density function $f_{(T,X)}(t, x)$ from Eq. (17) with the product $f_T(t)f_X(x)$ of the probability density function $f_T(t)$ from Eq. (12) and the probability density function $f_X(x)$ from Eq. (15), it is straightforward to see that

$$f_{(T,X)}(t,x) \neq f_T(t)f_X(x), \quad t,x > 0. \tag{18}$$

Relation (18) implies that the reaction time T and the reaction range X are *dependent* random variables. This dependence is diametric to the *conditional independence* of the reaction time T and the reaction range X, conditioned on the realizations of distances $\{D_i\}$, which is manifested by Eq. (9). Thus the random Poissonian structure of the distances $\{D_i\}$ induces a statistical dependence between the reaction time T and the reaction range X.

E. Conditional distributions

The conditional distribution of the reaction time T, conditioned on the realization of the reaction range X, is given by the tail distribution function

$$\Pr(T > t|X = x) = \frac{\int_t^\infty \Pr(T > s)\exp\{-\eta(x)s\}ds}{\int_0^\infty \Pr(T > s)\exp\{-\eta(x)s\}ds}, \tag{19}$$

for $t > 0$. Also, the conditional distribution of the reaction range X, conditioned to the realization of the reaction time T, is given by the tail distribution function

$$\Pr(X > x|T = t) = \frac{\int_x^\infty \exp\{-\eta(y)t\}\eta(y)\rho(dy)}{\int_0^\infty \exp\{-\eta(y)t\}\eta(y)\rho(dy)}, \tag{20}$$

for $x > 0$.

As noted at the end of Sec. II A, the RARE model can be viewed as a descendant of Blumen's unified donor-acceptor energy transfer model[28] and the donor-acceptor recombination model of Thomas *et al.*[29] In Refs. 28 and 29, the reaction time T was analyzed, and a counterpart of Eq. (10) was established (in the context of Bernoulli scattering of excitations over an N-site lattice). The analysis carried out in this section goes significantly beyond those of Refs. 28 and 29 as it addresses the marginal, joint, and conditional distributions of the reaction pair (T, X). Moreover, all results established in this section are valid in the context of general Poisson scattering of excitations over general metric spaces, a setting which allows for high variability and versatility while yielding closed-form analytic expressions.

IV. MONTE-CARLO SIMULATION

The stochastic analysis presented in Sec. III facilitates the construction of a numerical Monte-Carlo algorithm for the simulation of the reaction pair (T, X). The steps of the Monte-Carlo algorithm are as follows:

(1) Identify the inputs of the RARE model: the scattering function $\rho(x)$ and the reactivity function $\eta(x)$.
(2) Using the scattering function $\rho(x)$ and the reactivity function $\eta(x)$, numerically compute the integral

$$I(t,x) = \int_0^x \exp\{-\eta(y)t\}\eta(y)\rho(dy), \quad t,x \geq 0. \tag{21}$$

(3) Using the integral $I(t, x)$, numerically compute the cumulative distribution function of the reaction time

$$F(t) = 1 - \exp\left(-\int_0^t I(\tau,\infty)d\tau\right), \quad t \geq 0, \tag{22}$$

and then numerically compute its inverse function $F^{-1}(u)$ $(0 \leq u \leq 1)$.
(4) Using the integral $I(t, x)$, numerically compute the conditional cumulative distribution function of the reaction range

$$G(x;t) = \frac{I(t,x)}{I(t,\infty)}, \quad x \geq 0, \tag{23}$$

and then numerically compute its inverse function $G^{-1}(u; t)$ $(0 \leq u \leq 1)$.
(5) Generate a pair (U_1, U_2) of independent random variables, which are uniformly distributed over the unit interval, and generate the reaction pair (T, X) via

$$T = F^{-1}(U_1) \text{ and } X = G^{-1}(U_2; F^{-1}(U_1)). \tag{24}$$

The third step of the Monte-Carlo algorithm is based on Eq. (10), the fourth step is based on Eq. (20), and in the fifth step we applied the following basic simulation principle:[35] If $\Phi(x)$ $(x \geq 0)$ is the cumulative distribution function of a positive-valued random variable ξ, and if U is a random variable which is uniformly distributed over the unit interval (and independent of ξ), then the random variable $\Phi^{-1}(U)$ is equal in law to the random variable ξ.

The Monte-Carlo algorithm can be further used to simulate *random walks* whose dynamics are governed by the RARE model. To that end assume that the scattering of the excitations is spatially homogeneous. This homogeneity implies that the Poissonian structure of the distances $\{D_i\}$ is invariant with respect to the position P of the reaction center. Now consider the following reaction-based propagation scheme: if the reaction center reacted with excitation i, then the center jumps from its initial position P to the position of excitation i, P_i, and thereafter the process starts anew. This propagation scheme generates a renewal continuous time random walk (CTRW),[36–38] whose law of motion is as follows:

(1) Initiate from an arbitrary position P_0 at time t_0, and simulate a reaction pair (T, X).
(2) At time $t_1 = t_0 + T$ move to a point P_1 which is uniformly distributed on a sphere with radius X centered at the point P_0.
(3) Set $t_0 := t_1$ and $P_0 := P_1$, and go back to step (1).

The reaction time T is the CTRW's generic waiting time, and the reaction range X is CTRW's generic jump length. We emphasize that since the reaction time T and the reaction range X are *dependent* random variables, the RARE model induces CTRWs with *coupled* waiting times and jump lengths.[39–41]

V. POWER-LAW INPUTS

To illustrate the results established so far we consider now the example of the RARE model with power-law inputs. Specifically, we consider both the scattering function and the reactivity function to be power-laws

$$\rho(x) = ax^{\alpha} \text{ and } \eta(x) = bx^{-\beta}, \quad x > 0 \tag{25}$$

whose coefficients a and b as well as the exponents α and β are all positive parameters.

Power-law inputs occur naturally in many cases. If we scatter the excitations uniformly across d-dimensional Euclidean space using a spatially homogeneous Poisson process, then the scattering function $\rho(x)$ will be a power-law with exponent $\alpha = d$. Indeed, if the excitations are scattered uniformly across the d-dimensional Euclidean space, then the mean number of excitations present in a ball of radius x, quantified by the scattering function $\rho(x)$, is proportional to x^d. Moreover, in many fractal settings, which are abundant in the physical sciences, the scattering of the chemical agents is uniform across some fractal unbounded subset of the d-dimensional Euclidean space. In such fractal settings the scattering function $\rho(x)$ will be a power-law with exponent α that equals the fractal dimension of the underlying fractal subset.[42] On the other hand, power-law decays of physical and chemical interactions as a function of the distance between the interacting elements are prevalent in the physical sciences, the best known examples being Newton's law of gravitation, Coulomb's law, or the van der Waals law.

We turn now to describe the statistical behavior of the RARE model with power-law inputs. For the RARE model to be well defined, the exponent α of the scattering function must be smaller than the exponent β of the reactivity function, and thus we assume that $\alpha < \beta$.

A. Reaction time

From Eq. (10), we obtain that the tail distribution function of the reaction time T is given by

$$\Pr(T > t) = \exp(-c_1 t^{\alpha/\beta}), \quad t \geq 0, \tag{26}$$

where the precise value of the coefficient c_1 is given by $c_1 = \Gamma(1 - \alpha/\beta)ab^{\alpha/\beta}$. The tail distribution function of Eq. (26) characterizes a *stretched exponential* law.[43–45] The hazard rate of the reaction time T is a power-law with a negative exponent equal to $-(1 - \alpha/\beta)$, and the moments of the reaction time T are given by

$$\mathbf{E}[T^m] = \Gamma(1 + m\beta/\alpha)c_1^{-m\beta/\alpha}, \quad m > 0. \tag{27}$$

We emphasize that although the reaction time T has convergent moments of all orders, its moment generating function is divergent: $\mathbf{E}[\exp(\theta T)] = \infty$ for all $\theta > 0$. This statistical behavior of the stretched exponential distribution, convergent moments and divergent moment generating function, implies that the reaction time T displays a form of randomness which Mandelbrot categorized as "borderline randomness."[46] Another well-known probability distribution displaying such borderline randomness is the log-normal distribution.[47]

B. Reaction range

Evaluating Eq. (16), while applying a moment-expansion of the Laplace transform of the reaction time T, we obtain that the tail distribution function of the reaction range X is given by the power-expansion

$$\Pr(X > x) = a\alpha \sum_{m=1}^{\infty} (-1)^{m+1} \frac{b^m \mathbf{E}[T^m]}{m!(m\beta - \alpha)} \frac{1}{x^{m\beta-\alpha}} \tag{28}$$

with $x > 0$. In turn, Eq. (28) implies that the asymptotic behavior of the tail distribution function of the reaction range X is given by

$$\Pr(X > x) \approx \frac{c_2}{x^{\beta-\alpha}}, \quad x \to \infty, \tag{29}$$

where the coefficient c_2 is given by $c_2 = [\Gamma(1 + \beta/\alpha)a^{1-\beta/\alpha}]/[\Gamma(1 - \alpha/\beta)(\beta - \alpha)]$. The tail asymptotics of Eq. (29) characterize an *asymptotically Paretian* distribution with exponent $\beta - \alpha$.[30,48–50] The moments of the reaction range X are convergent, i.e., $\mathbf{E}[X^m] < \infty$, if and only if the exponent m is in the range $0 < m < \beta - \alpha$. In particular, the reaction range X has a convergent mean if and only if $1 + \alpha < \beta$. This statistical behavior of asymptotically Paretian distributions, convergent moments only up to a given order, implies that the reaction range X displays a form of randomness which Mandelbrot categorized as "wild randomness."[46,51] For a recent treatment of the "categorization of randomness" see Ref. 52.

C. Conditional distribution

From Eq. (20), we obtain that the conditional distribution of the reaction range X, conditioned on the realization of the reaction time T, is given by the tail distribution function

$$\Pr(X > x|T = t) = \int_0^{tbx^{-\beta}} \frac{\exp(-u)u^{-\alpha/\beta}}{\Gamma(1 - \alpha/\beta)} du, \quad x > 0. \tag{30}$$

Note that the integrand on the right-hand side of Eq. (30) is the probability density function of a *Gamma distribution* with exponent $1 - \alpha/\beta$. Thus, if we set ξ to be a Gamma-distributed random variable with exponent $1 - \alpha/\beta$, we obtain that $\Pr(X > x|T = t) = \Pr(\xi \leq tbx^{-\beta})$. Equation (30) further implies that the asymptotic behavior of the conditional tail distribution function of the reaction range X is given by

$$\Pr(X > x|T = t) \approx c_3 \frac{t^{1-\alpha/\beta}}{x^{\beta-\alpha}}, \quad x \to \infty, \tag{31}$$

where $c_3 = b^{1-\alpha/\beta}/\Gamma(2 - \alpha/\beta)$. As in the case of Eq. (29), the tail asymptotics of Eq. (31) characterize an asymptotically Paretian distribution with exponent $\beta - \alpha$. Consequently, also

with any given realization of the reaction time T, the reaction range X displays wild randomness.

VI. THERMODYNAMIC LIMIT

In this section, we explore the statistical behavior of the RARE model as the *concentration* of the excitations is increased to infinity. Specifically, we increase by n-fold the concentration of the excitations, and examine the limiting statistical behavior of the reaction time and the reaction range in the limit $n \to \infty$.

An n-fold increase of the concentration of the excitations results in replacing the "original" scattering function $\rho(x)$ by the "concentrated" scattering function $\rho_n(x) = n\rho(x)$. Note that an n-fold increase of the concentration of the excitations does not affect the reactivity function $\eta(x)$. In this section, we denote by (T_n, X_n) the reaction pair corresponding to a RARE model with scattering function $\rho_n(x)$ and reactivity function $\eta(x)$. Moreover, we set

$$\lambda = \int_0^\infty \eta(x)\rho(dx) \tag{32}$$

and assume that the integral on the right-hand side of Eq. (32) is convergent.

We introduce the scaled reaction time $S_n = nT_n$. A stochastic limit analysis detailed in Appendix E asserts that the joint probability density function of the reaction pair (S_n, X_n) attains the limit

$$\lim_{n\to\infty} f_{(S_n,X_n)}(s,x) = \exp(-\lambda s)\eta(x)\rho'(x), \quad s,x>0. \tag{33}$$

Namely, the reaction pair (S_n, X_n) converges in law (as $n \to \infty$) to a limiting random vector (S_∞, X_∞), whose distribution is governed by the joint probability density function

$$f_{(S_\infty,X_\infty)}(s,x) = \lambda \exp(-\lambda s)\frac{1}{\lambda}\eta(x)\rho'(x), \quad s,x>0. \tag{34}$$

Equation (34) implies the following:

(1) The scaled reaction time S_n converges in law to a stochastic limit S_∞, which is *exponentially distributed* with rate λ and density function

$$f_{S_\infty}(s) = \lambda \exp(-\lambda s), \quad s>0. \tag{35}$$

(2) The reaction range X_n converges in law to a stochastic limit X_∞ whose distribution is governed by the probability density function

$$f_{X_\infty}(x) = \frac{1}{\lambda}\eta(x)\rho'(x), \quad x>0. \tag{36}$$

(3) The joint probability density function of the random vector (S_∞, X_∞) equals the product of its corresponding marginal probability density functions

$$f_{(S_\infty,X_\infty)}(s,x) = f_{S_\infty}(s)f_{X_\infty}(x), \quad s,x>0, \tag{37}$$

and hence the stochastic limits S_∞ and X_∞ are *independent* random variables.

The stochastic limit result established in this section gets us "all around the circle." Indeed, in the preliminary analysis of Sec. II we obtained that the conditional distribution of the reaction time T, conditioned on the realizations of distances $\{D_i\}$, is exponential and independent of the reaction range X. However, in Sec. III we found that the distribution of the reaction time T is general and is tightly coupled to the distribution of the reaction range X. Thus, the random Poissonian structure of the distances $\{D_i\}$ shifts the distribution of the reaction time T from exponential to general, and induces a dependence between the reaction time T and the reaction range X. In this section, we established that in the infinite concentration limit $n \to \infty$ the original statistical structure is recovered: (i) the exponential distribution is restored, as the stochastic limit S_∞ of the scaled reaction time is exponentially distributed; and (ii) the independence is restored, as the stochastic limit S_∞ of the scaled reaction time and the stochastic limit X_∞ of the reaction range are independent random variables.

To illustrate the stochastic limit result established in this section consider the example of power law scattering functions studied in Sec. V and exponential reactivity functions

$$\rho(x) = ax^\alpha \text{ and } \eta(x) = b\exp(-\beta x), \quad x>0, \tag{38}$$

where the coefficients a and b as well as the parameters α and β, are all positive. In this example, the stochastic limit S_∞ is exponentially distributed with rate

$$\lambda = ab\frac{\Gamma(1+\alpha)}{\beta^\alpha}, \tag{39}$$

and the stochastic limit X_∞ is *Gamma distributed* with probability density function

$$f_{X_\infty}(x) = \frac{\beta^\alpha}{\Gamma(\alpha)}\exp(-\beta x)x^{\alpha-1}, \quad x>0. \tag{40}$$

Note that the Gamma probability density function of Eq. (40) is (i) unbounded and monotone decreasing in the exponent range $\alpha < 1$; (ii) bounded and monotone decreasing at the exponent value $\alpha = 1$, in which case X_∞ is exponentially distributed with mean $1/\beta$; (iii) bounded and unimodal, with mode $x_* = (\alpha - 1)/\beta$, in the exponent range $\alpha > 1$.

VII. CONCLUSION

In this paper, we presented a general spatio-chemical stochastic model for generalized RARE relaxation in complex disordered systems. The RARE model considers a collection of excitations which are randomly scattered around a reaction center in some general embedding metric space. The RARE model has two input quantities: (i) the scattering function $\rho(x)$ quantifying the scattering intensity of the excitations around the reaction center as a function of the distance $(x > 0)$ from the center; (ii) the reactivity function $\eta(x)$ quantifying the reaction rate between the excitations and the reaction center as a function of the distance $(x > 0)$ from the center. The scattering and reactivity functions provide a straightforward intuitive description, as well as a precise mathematical formulation, of general relaxation processes in complex disordered systems.

The RARE model has two random outputs: (i) the reaction time T of its random relaxation process; (ii) the reaction range X of its random relaxation process. A detailed

stochastic analysis of the reaction pair (T, X) was carried out, yielding closed form results regarding the statistics of this pair: marginal distributions, joint distribution, and conditional distributions. The results established further led to a Monte-Carlo algorithm for the simulation of the model's random relaxation process, and to the thermodynamic of the RARE model. In addition, we investigated in detail the case of power-law inputs, which were shown to yield stretched exponential relaxation patterns and asymptotically Paretian relaxation ranges.

The RARE model is a compact and transparent stochastic approach to non-exponential relaxation processes. On the one hand the model's inputs are both intuitively clear and mathematically precise and are directly related to the physical properties of the system considered. On the other hand the model is robust and versatile, as its general mathematical formulation accommodates diverse physical settings. We therefore expect the RARE model to be useful to a wide range of applications in the physical sciences and beyond.

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APPENDIX A: THE DISTRIBUTION OF THE REACTION TIME *T*

Equation (1) implies that the tail distribution function of the reaction time T is given by

$$\Pr(T > t) = \Pr\left(\min_i\{T_i\} > t\right) = \Pr\left(\bigcap_i\{T_i > t\}\right). \tag{A1}$$

After conditioning with respect to the distances $\{D_i\}$, this is equal to

$$\Pr(T > t) = \mathbf{E}\left[\Pr\left(\bigcap_i\{T_i > t\}|\{D_i\}\right)\right]. \tag{A2}$$

Using the assumptions of the RARE model and Eq. (4), we have

$$\begin{aligned}\Pr(T > t) &= \mathbf{E}\left[\prod_i \Pr(T_i > t|D_i)\right] \\ &= \mathbf{E}\left[\prod_i \exp\{-\eta(D_i)t\}\right].\end{aligned} \tag{A3}$$

With Campbell's theorem of the theory of Poisson processes (see Sec. 3.2 in Ref. 24),

$$\Pr(T > t) = \exp\left(-\int_0^\infty [1 - \exp\{-\eta(x)t\}]\rho(dx)\right). \tag{A4}$$

Equations (A1) to (A4) prove Eq. (10).

APPENDIX B: THE DISTRIBUTION OF THE REACTION PAIR (*T, X*)

In what follows we set

$$E(t, \theta, x) = \mathbf{E}[\exp\{-tR(0) - \theta R(x)\}], \quad t, \theta, x \geq 0. \tag{B1}$$

Equation (5) implies that

$$\begin{aligned}tR(0) + \theta R(x) = \sum_i &(t\eta(D_i)\mathbf{I}(D_i \leq x) \\ &+ (t + \theta)\eta(D_i)\mathbf{I}(D_i > x)),\end{aligned} \tag{B2}$$

and hence Eq. (B1) further implies that

$$\begin{aligned}E(t, \theta, x) = \mathbf{E}\Big[\prod_i &\exp(-t\eta(D_i)\mathbf{I}(D_i \leq x) \\ &- (t + \theta)\eta(D_i)\mathbf{I}(D_i > x))\Big].\end{aligned} \tag{B3}$$

Consequently, Campbell's theorem of the theory of Poisson processes (see Sec. 3.2 in Ref. 24) implies that

$$E(t, \theta, x) = \exp\{-\Psi(t, \theta, x)\}, \tag{B4}$$

where

$$\begin{aligned}\Psi(t, \theta, x) &= \int_0^\infty [1 - \exp\{-t\eta(s)\mathbf{I}(s \leq x) \\ &\quad - (t + \theta)\eta(s)\mathbf{I}(s > x)\}]\rho(ds) \\ &= \int_0^x [1 - \exp\{-t\eta(s)\}]\rho(ds) \\ &\quad + \int_x^\infty [1 - \exp\{-(t + \theta)\eta(s)\}]\rho(ds).\end{aligned} \tag{B5}$$

Applying conditioning with respect to the distances $\{D_i\}$ and using Eq. (8), we obtain that the joint tail distribution function of the reaction pair (T, X) is given by

$$\begin{aligned}\Pr(T > t, X > x) &= \mathbf{E}[\Pr(T > t, X > x|\{D_i\})] \\ &= \mathbf{E}\left[\exp\{-tR(0)\}\frac{R(x)}{R(0)}\right], \quad t, x \geq 0.\end{aligned} \tag{B6}$$

Differentiating this expression with respect to the variable t yields

$$\frac{\partial}{\partial t}\Pr(T > t, X > x) = -\mathbf{E}[\exp\{-tR(0)\}R(x)]. \tag{B7}$$

Conversely, differentiating Eq. (B1) with respect to the variable θ yields

$$\frac{\partial}{\partial \theta}E(t, \theta, x) = -\mathbf{E}[\exp\{-tR(0) - \theta R(x)\}R(x)]. \tag{B8}$$

Combination of Eqs. (B7) and (B8) yields

$$\frac{\partial}{\partial t}\Pr(T > t, X > x) = \frac{\partial}{\partial \theta}E(t, \theta, x)\Big|_{\theta=0}. \tag{B9}$$

Now, Eq. (B4) implies that

$$\frac{\partial}{\partial \theta}E(t, \theta, x)\Big|_{\theta=0} = -\exp\{-\Psi(t, 0, x)\}\ \frac{\partial}{\partial \theta}\Psi(t, \theta, x)\Big|_{\theta=0}, \tag{B10}$$

and Eq. (B5) further implies that

$$\Psi(t,0,x)=\int_0^\infty [1-\exp\{-t\eta(s)\}]\rho(ds) \tag{B11}$$

and

$$\frac{\partial}{\partial\theta}\Psi(t,\theta,x)\Big|_{\theta=0}=\int_x^\infty \exp\{-t\eta(s)\}\eta(s)\rho(ds). \tag{B12}$$

Thus, combining Eqs. (B9)–(B12), we conclude that

$$\begin{aligned}-\frac{\partial}{\partial t}\Pr(T>t,X>x)&=\exp\left(-\int_0^\infty [1-\exp\{-t\eta(s)\}]\rho(ds)\right)\\&\quad\times\int_x^\infty \exp\{-t\eta(s)\}\eta(s)\rho(ds).\end{aligned} \tag{B13}$$

Finally, differentiating this last expression with respect to the variable x and using Eq. (10), we arrive at the desired result,

$$\frac{\partial^2}{\partial t\partial x}\Pr(T>t,X>x)=\Pr(T>t)\exp\{-t\eta(x)\}\eta(x)\rho'(x), \tag{B14}$$

such that Eq. (B14) proves Eq. (17).

APPENDIX C: THE MARGINAL DISTRIBUTIONS OF THE REACTION PAIR (*T*, *X*)

The probability density function of the reaction time T is attained by integrating the joint probability density function of the random pair (T, X) over the distance variable x,

$$f_T(t)=\int_0^\infty f_{(T,X)}(t,x)dx. \tag{C1}$$

With Eq. (17),

$$f_T(t)=\Pr(T>t)\int_0^\infty \exp\{-t\eta(x)\}\eta(x)\rho(dx),\quad t>0. \tag{C2}$$

Equations (C1) and (C2) prove Eq. (12). Note that the Laplace transform of the reaction time T is given by

$$\mathbf{E}[\exp(-\theta T)]=\int_0^\infty \exp(-\theta t)f_T(t)dt. \tag{C3}$$

Integration by parts yields

$$\mathbf{E}[\exp(-\theta T)]=1-\theta\int_0^\infty \exp(-\theta t)\Pr(T>t)dt, \tag{C4}$$

where $\theta\geq 0$.

The probability density function of the reaction range X is attained by integrating the joint probability density function of the random pair (T, X) over the time variable t,

$$f_X(x)=\int_0^\infty f_{(T,X)}(t,x)dt \tag{C5}$$

and, using Eq. (17),

$$f_X(x)=\left(\int_0^\infty \Pr(T>t)\exp\{-t\eta(x)\}dt\right)\eta(x)\rho'(x). \tag{C6}$$

With Eqs. (C3) and (C4) and $\theta=\eta(x)$, we find

$$f_X(x)=(1-\mathbf{E}[\exp\{-\eta(x)T\}])\rho'(x),\quad x>0. \tag{C7}$$

Equations (C5)–(C7) prove Eq. (15).

APPENDIX D: THE CONDITIONAL DISTRIBUTIONS OF THE REACTION PAIR (*T*, *X*)

The conditional probability density function of the reaction time T, conditioned to the realization of the reaction range X, is given by

$$f_{T|X=x}(t)=\frac{f_{(T,X)}(t,x)}{f_X(x)}. \tag{D1}$$

With Eqs. (15) and (17),

$$\begin{aligned}f_{T|X=x}(t)&=\frac{\Pr(T>t)\exp\{-t\eta(x)\}\eta(x)\rho'(x)}{\left(\int_0^\infty \Pr(T>\tau)\exp\{-\tau\eta(x)\}d\tau\right)\eta(x)\rho'(x)}\\&=\frac{\Pr(T>t)\exp\{-t\eta(x)\}}{\int_0^\infty \Pr(T>\tau)\exp\{-\tau\eta(x)\}d\tau}.\end{aligned} \tag{D2}$$

In turn, Eqs. (D1) and (D2) imply Eq. (19).

The conditional probability density function of the reaction range X, conditioned to the realization of the reaction time T, is given by

$$f_{X|T=t}(x)=\frac{f_{(T,X)}(t,x)}{f_T(t)}. \tag{D3}$$

Using Eqs. (12) and (17), we see that

$$\begin{aligned}f_{X|T=t}(x)&=\frac{\Pr(T>t)\exp\{-t\eta(x)\}\eta(x)\rho'(x)}{\Pr(T>t)\int_0^\infty \exp\{-\eta(x)t\}\eta(x)\rho(dx)}\\&=\frac{\exp\{-t\eta(x)\}\eta(x)\rho'(x)}{\int_0^\infty \exp\{-\eta(x)t\}\eta(x)\rho(dx)}.\end{aligned} \tag{D4}$$

In turn, Eqs. (D3) and (D4) imply Eq. (20).

APPENDIX E: THE LIMIT DISTRIBUTION OF THE REACTION PAIR (S_n, X_n)

The scaling $S_n=nT_n$ implies the following connection between the joint cumulative distribution function of the reaction pair (S_n, X_n) and the joint cumulative distribution function of the reaction pair (T_n, X_n),

$$\Pr(S_n\leq s,X_n\leq x)=\Pr\left(T_n\leq\frac{s}{n},X_n\leq x\right),\quad s,x>0. \tag{E1}$$

Consequently, differentiating this expression with respect to the variables s and x yields the following connection between the joint probability density function of the reaction pair (S_n, X_n) and the joint probability density function of the reaction pair (T_n, X_n),

$$f_{(S_n,X_n)}(s,x)=f_{(T_n,X_n)}\left(\frac{s}{n},x\right)\frac{1}{n}. \tag{E2}$$

Using Eq. (17) with the concentrated scattering function $\rho_n(x)$, we arrive at

$$f_{(S_n,X_n)}(s,x)=\Pr\left(T_n>\frac{s}{n}\right)\exp\left(-\frac{s}{n}\eta(x)\right)\frac{\eta(x)\rho_n'(x)}{n}. \tag{E3}$$

With the definition of the concentrated scattering function $\rho_n(x)$, this implies

$$f_{(S_n,X_n)}(s,x) = \Pr\left(T_n > \frac{s}{n}\right) \exp\left(-\frac{s}{n}\eta(x)\right) \eta(x)\rho'(x). \tag{E4}$$

Equations (E2)–(E4) lead us to conclude that

$$\lim_{n\to\infty} f_{(S_n,X_n)}(s,x) = \left[\lim_{n\to\infty} \Pr\left(T_n > \frac{s}{n}\right)\right] \eta(x)\rho'(x) \tag{E5}$$

with $s, x > 0$. Now, using Eq. (10) with the concentrated scattering function $\rho_n(x)$ we find

$$\lim_{n\to\infty} \Pr\left(T_n > \frac{s}{n}\right)$$
$$= \lim_{n\to\infty} \exp\left(-\int_0^\infty \left[1 - \exp\left(-\eta(x)\frac{s}{n}\right)\right] \rho_n(dx)\right). \tag{E6}$$

With the definition of the concentrated scattering function $\rho_n(x)$ this leads to

$$\lim_{n\to\infty} \Pr\left(T_n > \frac{s}{n}\right)$$
$$= \exp\left(-\int_0^\infty \left[\lim_{n\to\infty} \frac{1 - \exp\left(-\eta(x)t/n\right)}{1/n}\right] \rho(dx)\right). \tag{E7}$$

Finally, with de l'Hospital's rule and Eq. (32) we obtain

$$\lim_{n\to\infty} \Pr\left(T_n > \frac{s}{n}\right)$$
$$= \exp\left(-\int_0^\infty [\eta(x)t]\rho(dx)\right) = \exp(-\lambda t). \tag{E8}$$

Combining together the limits of Eqs. (E4) and (E8) proves Eq. (33).

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