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Characterization of the non-Arrhenius behavior of supercooled liquids by modeling nonadditive stochastic systems

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The characterization of the formation mechanisms of amorphous solids is a large avenue for research, since understanding its non-Arrhenius behavior is challenging to overcome. In this context, we present one path toward modeling the diffusive processes in supercooled liquids near glass transition through a class of nonhomogeneous continuity equations, providing a consistent theoretical basis for the physical interpretation of its non-Arrhenius behavior. More precisely, we obtain the generalized drag and diffusion coefficients that allow us to model a wide range of non-Arrhenius processes. This provides a reliable measurement of the degree of fragility of the system and an estimation of the fragile-to-strong transition in glass-forming liquids, as well as a generalized Stokes-Einstein equation, leading to a better understanding of the classical and quantum effects on the dynamics of nonadditive stochastic systems.

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I. INTRODUCTION

The dynamic response of a wide class of materials can be achieved using the so-called Arrhenius law [1–5]. Basically, it consists of an exponential decay with the inverse of the temperature characterized by the so-called temperature independent activation energy [1–3]. The search for a physical interpretation of the activation energy established the fundamentals of the transition state theory [6–8] since it associates an Arrhenius-like behavior with diffusive processes in several systems [4,9–12].

However, from the development of new technologies and advances in materials preparation techniques, a wide variety of new compounds could be synthesized, leading to the improvement of experimental techniques for the study of chemical reactions and diffusive processes. In this scenario, several systems have revealed deviations from Arrhenius behavior, evidenced through the temperature dependence of the activation energy [1]. In recent years, the characterization of non-Arrhenius behaviors has received considerable attention, since it was observed in water-type models SPC/E (extended simple point charge) [13,14], food systems [15], diffusivity in supercooled liquids near glass transition [2,10,11,16,17], chemical reactions [8,18,19], and several biological processes [20,21]. Therefore, modeling these non-Arrhenius systems is a large avenue for research and an actual challenge to overcome.

The non-Arrhenius behaviors manifest themselves as concave curves (sub-Arrhenius behavior), associated with nonlocal quantum effects [8,19,22], or convex curves (super-Arrhenius behavior), associated with the predominance of classical transport phenomena [3,20,22,23]. Despite much effort by the scientific community, there are only a few phenomenological relationships proposed to model non-Arrhenius processes, such as the Vogel-Tamman-Fulcher equation [24–26] and the Aquilanti-Mundim d-Arrhenius model [3,19,20,22,23,27,28]. Other phenomenological expressions have recently been proposed [5,10,17]. However, there is a need to establish a wide class of equations that characterize non-Arrhenius processes in a consistent theoretical basis for the physical interpretation of the characteristic non-Arrhenius behavior of several diffusive processes.

Nevertheless, the Aquilanti-Mundim equation can be derived from the stationary process of the nonlinear Fokker-Planck equation, and the diffusivity dependence with the temperature is consistent with experimental results [5]. Nonlinear Fokker-Planck equations, especially those whose stationary solutions maximize nonadditive entropies [29], such as the Tsallis entropy [30], have been successfully employed for modeling non-Markovian processes [31,32], anomalous diffusion [33,34], astrophysical systems [35], sunspots [36], and pitting corrosion [37], suggesting that this class of equations can also be an alternative way to describe the non-Arrhenius behavior of nonadditive stochastic systems.

In this context, we show in this paper a class of nonhomogeneous continuity equations whose generalized coefficient allows the modeling of a wide range of non-Arrhenius processes. We model the characteristic super-Arrhenius behavior of diffusivity and viscosity in supercooled liquids, determining a characteristic threshold temperature associated with the discontinuities in its dynamic properties, such as the viscosity and the activation energy. In addition, we define a generalized exponent that characterizes the non-Arrhenius process and serves as an indicator of the level of fragility in glass-forming systems, whereas the threshold temperature

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indicates a fragile-to-strong transition, the general behavior of metallic glass-forming liquids [38]. Our model also derives a generalized version for the Stokes-Einstein equation, where we obtain a characteristic temperature independent behavior (at low temperatures) for sub-Arrhenius processes, and a sudden death behavior around the threshold temperature for super-Arrhenius processes. Our results pave the way for the characterization of the breakdown of the standard Stokes-Einstein relation [39–42], mainly in supercooled liquids [43], providing one path toward understanding the dynamic evolution of non-Arrhenius processes, leading to the establishment of a theoretical interface between a macroscopic and microscopic perspective of the matter through a nonequilibrium statistical mechanics.

II. GENERALIZED REACTION-DIFFUSION MODEL

Let us consider a concentration $\rho(r,t)$ of a substance measured in volume V at time t; the total amount of substance for the same volume is given by the nonhomogeneous continuity equation. In this context, we propose a generalized model, based on a nonhomogeneous continuity equation, which refers to dissipative reaction-diffusion processes, through the following conditions:

(i) $f(r,t) = \vec{\nabla}\cdot\vec{v}(r,t)$ is a volumetric density per unit time associated with dissipative processes and $\vec{v}(r,t)$ is a field of nonzero divergence, which can be understood as a dissipation field, associated with these dissipative reaction-diffusion processes, by analogy to a source effect.

(ii) $\vec{v}(r,t) = -\kappa_m^{-1}\rho^m\vec{\nabla}\phi$, where κ_m is a positive constant parametrized by the exponent m. The potential ϕ is analogous to a generalized chemical potential μ, associated with the molar energy of a chemical reaction, in a reaction-diffusion model.

(iii) For the steady state $\vec{v}(r,t) \to \vec{v}_S(r)$, which is a field of zero divergence.

(iv) $\vec{J} = -D(r,t;\rho)\vec{\nabla}\rho$ is a diffusion flux, for a generalized version of Fick's first law [12,44], in which $D(r,t;\rho)$ is a generalized diffusion coefficient.

(v) $D(r,t;\rho) = (\Gamma/2)\rho^{n-1}$ [29], where Γ is a positive definite parameter, related to a class of nonlinear equations associated with anomalous diffusive processes [29,45]. This condition establishes the connection between nonlinear Fokker-Planck equations and nonadditive entropic forms [29,45]. In this context, nonlinear Fokker-Planck equations can be arranged into classes associated with these entropic forms and their corresponding stationary state, which is one path toward the characterization of the non-Arrhenius behavior of nonadditive stochastic systems.

In this circumstance, the nonhomogeneous continuity equation becomes a particular class of nonlinear Fokker-Planck equations [29] whose nonlinearity of the generalized drag coefficient involves the information of the dissipative or exchange processes, such as phase transitions or chemical reactions. From these conditions we obtain an alternative way to describe the non-Arrhenius behavior of the diffusion processes of nonadditive stochastic systems such as supercooled liquids, from a consistent theoretical basis. To ensure that the diffusion coefficient, $D(r,t;\rho)$, defined in condition (v) is proportional to the concentration $\rho(r,t)$, we consider the particular case $n = 2$. Thus, we obtain the nonhomogeneous continuity equation

$$\frac{\partial\rho(r,t)}{\partial t} = \kappa_m^{-1}\vec{\nabla}\cdot[(\vec{\nabla}\phi)\rho^m] + \frac{\Gamma}{2}\nabla^2[\rho^2], \qquad (1)$$

where Eq. (1) is a class of nonlinear Fokker-Planck equations, whose solutions compose a class of rapidly decreasing functions [46] that maximize nonadditive entropies, such as the Tsallis entropy [30], since this guarantees the possibility of fundamental solutions for the diffusion equation.

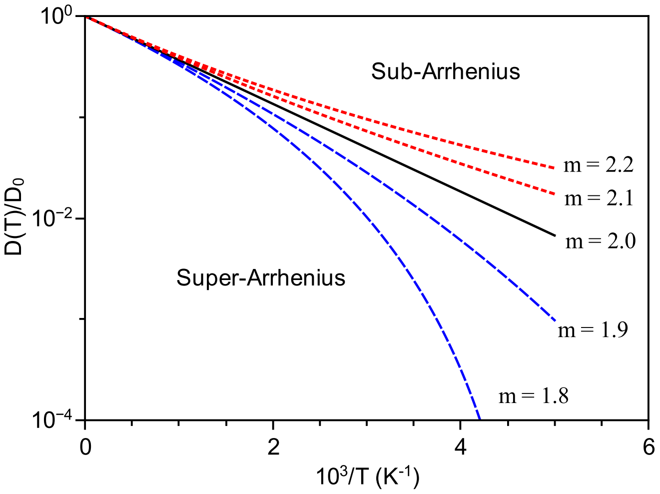

FIG. 1. Monolog plot of the diffusivity as a function of the reciprocal temperature. The curves $m > 2$ (dotted red lines) characterize a class of sub-Arrhenius processes, while the curves $m < 2$ (dashed blue lines) characterize a class of super-Arrhenius processes. The $m = 2$ curve (solid line) corresponds to the usual Arrhenius plot. The curves were simulated for the $E/k_B = 1000$ K condition

III. DIFFUSIVITY, VISCOSITY, AND FRAGILITY OF GLASS-FORMING LIQUIDS

The characterization of diffusivity and viscosity in supercooled liquids is effective to understand the glass transition and the formation mechanisms of amorphous solids. From the stationary solution of Eq. (1) and condition (v), for $n = 2$, the dependence of the diffusion coefficient with the temperature can be written as

$$D(T) = D_0\left[1-(2-m)\frac{E}{k_BT}\right]^{\frac{1}{2-m}}, \qquad (2)$$

where $D_0 = \Gamma C_0$ (C_0 is a normalization constant of the stationary concentration), $E = -\int\vec{\nabla}\phi\cdot dr$ is a generalized energy, and $C_0^{2-m}\kappa_m\Gamma = k_BT$ [47,48]. From Eq. (2), the Arrhenius standard behavior is recovered when the coefficient $m \to 2$; then the energy E, in this limit, corresponds to a temperature independent energy.

Figure 1 shows the diffusivity of a supercooled liquid as a function of the reciprocal temperature. Under the condition $m < 2$ the proposed model encompasses a class of super-Arrhenius diffusive processes, associated with the predominance of classical transport phenomena [3,20,22,23], predominantly according to experimental reports [5,10,16,17]. In

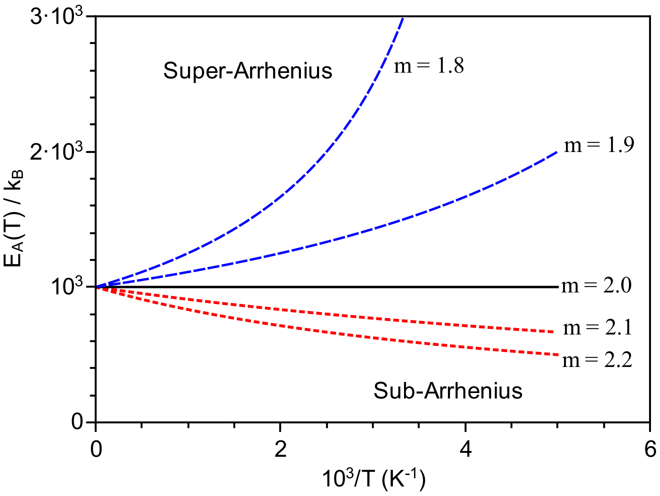

FIG. 2. The activation energy as a function of the reciprocal temperature. The curves $m > 2$ (dotted red lines), from which the activation energy is a decreasing function of the reciprocal temperature, characterize a class of sub-Arrhenius processes, while the increasing curves $m < 2$ (dashed blue lines) characterize a class of super-Arrhenius processes. In addition, the $m = 2$ curve (solid line) corresponds to the Arrhenius activation energy, characterized by a temperature independent behavior. The curves were simulated using the scale factor $E/k_B = 1000$ K.

addition, the model also covers a wide class of sub-Arrhenius diffusive processes, characterized by the condition $m > 2$, associated with nonlocal quantum effects [8,19,22], and less sensitive to the exponent variations than the super-Arrhenius processes.

It is also possible to verify the existence of a threshold temperature for super-Arrhenius processes, from which the diffusivity goes to zero, given by

$$T_t = \frac{(2-m)E}{k_B}. \tag{3}$$

From Eq. (2), using the slope of $\log(D(T)/D_0)$ as a function of T^{-1} as shown in Fig. 1, we obtain the activation energy temperature dependence as

$$E_A(T) = \frac{E}{1-(2-m)\frac{E}{k_B T}}\ , \tag{4}$$

the main feature of non-Arrhenius processes. Furthermore, from Eq. (4), for $m \to 2$ the activation energy achieves a temperature independent behavior $E_A(T) \to E$ corresponding to the Arrhenius law, as previously mentioned.

Figure 2 shows the activation energies, corresponding to the diffusivity curves presented in Fig. 1, calculated from Eq. (4). The activation energy is an increasing function of the reciprocal temperature for sub-Arrhenius processes and decreasing for super-Arrhenius processes. In addition, for the super-Arrhenius processes, when the threshold temperature, Eq. (3), is achieved the activation energy diverges to infinity, indicating that this temperature is related to the viscosity divergence in the glass transition.

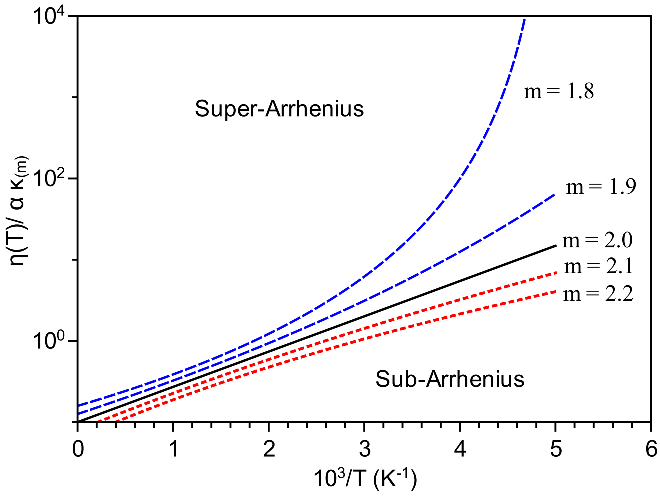

FIG. 3. The viscosity as a function of the reciprocal temperature. The curves $m > 2$ (dotted red lines) characterize a class of sub-Arrhenius models for viscosity, while the curves $m < 2$ (dashed blue lines) characterize a class of super-Arrhenius models. The $m = 2$ curve (solid line) corresponds to the Arrhenius model for the viscosity.

From Eq. (1), we can define $\kappa_m^{-1}\rho^{m-1}$ as the generalized fluid mobility [47], which relates the drift velocity of the diffusive flux to the dissipative field $\vec{v}(r,t) = -\kappa_m^{-1}\rho^m\vec{\nabla}\phi$, as described in Sec. II. Given that the viscosity $\eta(T)$ is inversely proportional to the fluid mobility, its temperature dependence is given by

$$\eta(T) = \eta_\infty\left[1-(2-m)\frac{E}{k_B T}\right]^{\frac{1-m}{2-m}}, \tag{5}$$

where $\eta_\infty = \alpha\kappa_m C_0^{1-m}$ is the high temperature viscosity limit and α is a positive definite constant. From Eq. (5) the Arrhenius model from the viscosity is recovered for the limit case $m \to 2$.

Figure 3 shows the viscosity as a function of the reciprocal temperature. For super-Arrhenius processes ($m < 2$) the threshold temperature characterizes the regime from which the viscosity diverges to infinity. Thus, the threshold temperature, Eq. (3), serves as an indication of how close the system is to the glass transition region because it involves discontinuities in the dynamic properties, such as the activation energy, Eq. (4), and viscosity, Eq. (5). The glass-liquid transition occurs in a range of temperatures for which the viscosity assumes a large value, but still does not diverge. In most glass-forming liquids, the glass transition temperature is established at the viscosity reference value of 10^{12} Pa s; thus $T_t \leqslant T_g$.

This model can also be used to calculate the level of fragility M_η in glass-forming systems [38,49,50] by our exponent m as

$$M_\eta = \left(\frac{m-1}{2-m}\right)\left(\frac{1}{1-\frac{T_t}{T_g}}\right). \tag{6}$$

For the usual Arrhenius diffusive processes, the condition $m = 2$ characterizes a *strong* glass system, whereas for a wide class of super-Arrhenius diffusive processes the condition $m < 2$ characterizes a *fragile* glass [49,50]. A second important feature that arises from our model is the distinguishability between strong and fragile systems for super-Arrhenius processes ($m < 2$), since the further the glass transition temperature T_g is from the threshold temperature, Eq. (3), the more fragile the system will be. The proposed approach considers a dynamic interpretation for the glass transition since the ratio T_t/T_g in Eq. (6) is related to discontinuities in dynamic properties such as energy and viscosity, indicating a *fragile-to-strong transition* [38] usually found in some water and silica systems, which is possibly a general behavior of metallic glass-forming liquids [38], where an initially fragile supercooled liquid can be transformed into a strong liquid upon supercooling toward T_g.

In this context, the transition characterized by this threshold temperature differs from the freezing or crystallization transition, which is in the Ehrenfest classification a first-order phase transition [51], because it cannot characterize a transition between states in the thermodynamic equilibrium. Therefore, the dynamics around the glass transition region, characterized by Eq. (3), provides a measurement of how fragile a system is, establishing the theoretical basis for the understanding of the formation mechanisms of amorphous solids.

Furthermore, fragile-to-strong transitions have been related to an extensive configurational entropy for all nonzero temperatures [52]. This feature can be identified in our model given that nonadditive entropic forms can be extensive, depending on the characteristics of the system, because additivity and extensivity are distinct properties that coincide only in short-range interacting systems from which is applicable the Boltzmann-Gibbs entropy [53–55].

Moreover, a third remarkable result can be extracted from our model. The product between the generalized diffusion coefficient, Eq. (2), and the viscosity, Eq. (5), obtained from our generalized model for reaction-diffusion processes provides a generalized Stokes-Einstein relation for any non-Arrhenius diffusion process, given by

$$D\eta = \alpha k_B T\left[1 - (2-m)\frac{E}{k_B T}\right]. \tag{7}$$

Figure 4 shows the temperature dependence of the generalized Stokes-Einstein relation, Eq. (7), for different values of the coefficient m. For the super-Arrhenius diffusive processes ($m < 2$) the relation gives an estimate of the glass transition temperature, since the generalized diffusion coefficient, Eq. (2), goes to zero faster than the viscosity, Eq. (6), diverges to infinity. Thus, the region in which the product of the viscosity and the generalized diffusion coefficient, Eq. (7), goes to zero is equivalent to the threshold temperature of glass transition, Eq. (3). In addition, as demonstrated in Fig. 4, the usual form of the Stokes-Einstein relation is recovered from Eq. (7) under two conditions: (i) for any Arrhenius-like process ($m \to 2$) and (ii) for the condition $E \ll k_B T$, i.e., thermal fluctuations predominate in the process, to the detriment of the concentration gradient.

On the other hand, for the sub-Arrhenius diffusive processes, it is worth noting that, from the condition $E \gg k_B T$, the generalized Stokes-Einstein equation, Eq. (7), presents a temperature independent behavior, enabling the differentiation of the classical and quantum regimes, paving the way for the characterization of sub-Arrhenius processes through Eq. (7). This provides one path toward understanding the quantum effects in the dynamics of the nonadditive stochastic systems.

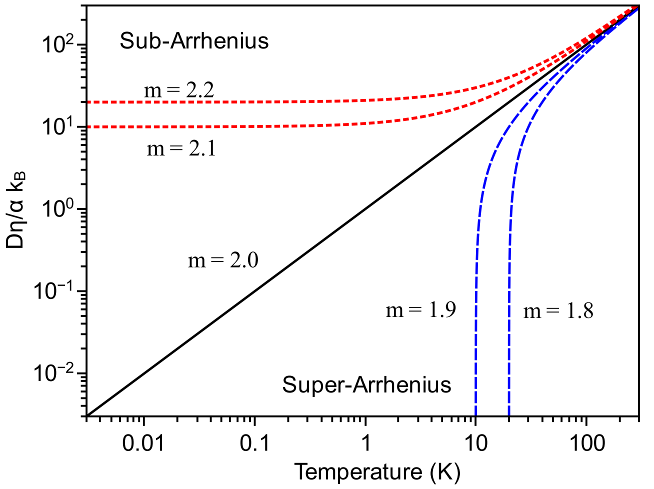

FIG. 4. The temperature dependence of the generalized Stokes-Einstein relation, Eq. (7), for different values of the coefficient m. For super-Arrhenius processes, $m < 2$ (dashed blue lines), the region in which the product between the viscosity and the generalized diffusion coefficient, Eq. (7), rapidly goes to zero is equivalent to the threshold temperature of glass transition, Eq. (3). For sub-Arrhenius processes, $m > 2$ (dotted red lines), from the condition $E \gg k_B T$ the generalized Stokes-Einstein equation presents a temperature independent behavior. The straight line corresponds to the usual form of the Stokes-Einstein relation, recovered for any Arrhenius-like process, $m = 2$ curve (solid line), and for the condition $E \ll k_B T$, that separates the super- and sub-Arrhenius regimes.

IV. CONCLUSIONS

In summary, our main result was to provide an alternative way to describe the non-Arrhenius behavior of diffusive processes in glass-forming liquids. Our model was characterized by a generalized exponent m that defines the class of non-Arrhenius processes and serves as an indicator of the degree of the fragility in these systems. In addition, we determine the threshold temperature, Eq. (3), from which the dynamic properties, such as the activation energy and viscosity, diverge and give us a reliable estimate of the degree of fragility, since the ratio T_t/T_g [Eq. (6)] indicates a fragile-to-strong transition, establishing the theoretical basis for understanding the intrinsic features of amorphous solids.

Also interesting is the realization of a generalized Stokes-Einstein equation, Eq. (7), which allows us to characterize the breakdown of the standard Stokes-Einstein relation in supercooled liquids. For sub-Arrhenius processes, the generalized relation presents a characteristic temperature independent behavior at low temperatures while, for the class of super-Arrhenius diffusive processes, it rapidly goes to zero around the threshold temperature. Moreover, the usual form of the Stokes-Einstein relation is recovered for any Arrhenius-like process and when the thermal fluctuations predominate in the process to the detriment of the concentration gradient ($E \ll k_B T$). Our results provide one path toward the differentiation of the super- and sub-Arrhenius processes, leading to a better understanding of the classical and quantum effects on the dynamics of nonadditive stochastic systems, paving the way for the characterization of the formation mechanisms of amorphous solids through the study of non-Arrhenius diffusive processes in these systems.

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