Relaxation in yield stress systems through elastically interacting activated events

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We study consequences of long-range elasticity in thermally assisted dynamics of yield stress materials. Within a 2d mesoscopic model we calculate the mean-square displacement and the dynamical structure factor for tracer particle trajectories. The ballistic regime at short time scales is associated with a compressed exponential decay in the dynamical structure factor, followed by a sub-diffusive crossover prior to the onset of diffusion. We relate this crossover to spatio-temporal correlations and thus go beyond established mean field predictions.

Introduction

Relaxation of the microscopic structure in glasses, and more generally in soft yield stress materials, is a topic of long standing interest and great complexity. Broad ranges of time, energy and length scales are involved, together with non-equilibrium aspects such as aging and a strong dependence on the sample preparation scheme. As a result, no unique scenario has emerged to describe the relaxation of density fluctuations in systems that, quenched from a liquid into a glassy (solid) state, still display internal dynamics strong enough to produce structural relaxation on a measurable time scale. The complexity of the relaxation is usually quantified by the manner in which it deviates from exponential. In many cases, stretching, corresponding to a broad distribution of relaxation times, is observed. However, the opposite situation of compressed relaxation (i.e., faster than exponential) has emerged in the last years as a new paradigm. In this work, we confirm through the numerical study of a simplified model that this behavior can result from thermally activated plastic events akin to the shear transformations observed in yield stress solids undergoing external deformation.

Background

Experimental analysis of relaxation processes has been carried out originally by dynamic light scattering (DLS) on colloid gels [1-3] and more recently by x-ray photon correlation spectroscopy (XPCS) in several soft-[4-6] and hard-amorphous systems [7]. A common observation is the decay of the dynamical structure factor as a compressed exponential in time t and scattering vector q.

$$f(q,t) \sim exp[-(t/\tau_f)^\gamma] \quad \text{with} \quad \tau_f \sim q^{-n}, \quad n \simeq 1 \quad \text{and shape parameter} \quad \gamma > 1$$

Both, faster than exponential correlations decay and ballistic dynamics contrasting the usual diffusive behavior ($\tau_f \sim q^{-2}$), were unexpected.

Originally, a heuristic explanation for the observed phenomena was based on the "syneresis" of a gel (local shrinking, inhomogeneity acts as a dipole force with a long range elastic effect) [1]. A mean-field approach [8,9] further encouraged the view that anomalous relaxation has its origin in elasticity effects, and introduced an important timescale in this analysis, namely θ the typical duration of an event. For a 3d system:

$$f(q,t) \sim e^{-a_1 (qt)^{3/2}} \quad \text{for} \quad t \ll \theta \qquad \text{and} \qquad f(q,t) \sim e^{-a_2 q^{3/2} t} \quad \text{for} \quad t \gg \theta$$

Later, a phenomenological continuous time random walk (CTRW) model with Lévy flights was introduced [3] to fit a crossover, with q, between compressed and non-compressed behaviors also observed in this experiments.

Observables

Mean square displacement $\langle \Delta r^2 \rangle$ averaged over number of tracers ($M \sim 2^{13}$) and sliding time windows. $\Delta r_i = |\boldsymbol{r}_i(t_0+t) - \boldsymbol{r}_i(t_0)|$

Dynamical structure factor (analogous to the experimental $f(q,t)$) $S(q,t) = \frac{1}{M} \left\langle \left[\sum_{n=1}^{M} \cos[\boldsymbol{q} \cdot (\boldsymbol{r}_n(t+t_0) - \boldsymbol{r}_n(t_0))] \right] \right\rangle_{t_0, |\boldsymbol{q}|=q}$

Elapsed time between two consecutive activations of the same site τ_{ev} and its distribution $\Psi(\tau_{ev})$

Absolute tracer displacement per unit time $u = |\boldsymbol{u}|$ and its distribution $P(u)$

Two time vectorial displacements autocorrelation function averaged over tracers and t_0 $\tilde{C}_{\boldsymbol{u}}(t) = \langle \boldsymbol{u}_{t_0} \cdot \boldsymbol{u}_{t_0+t} \rangle - \langle \boldsymbol{u}_{t_0} \rangle \cdot \langle \boldsymbol{u}_{t_0+t} \rangle$

Two time stress autocorrelation averaged over sites and t_0 $\tilde{C}_\sigma(t) = \langle \sigma_{t_0} \sigma_{t_0+t} \rangle - \langle \sigma_{t_0} \rangle \langle \sigma_{t_0+t} \rangle$

Yield stress *thermal* model

A 2d coarse-grained model with two main ingredients: thermally activated yield events (plastic rearrangements) and a long-range elastic response of the surrounding medium. We assume scalar quantities for local stresses $\sigma(\boldsymbol{r},t)$ and deformations $\epsilon^{pl}(\boldsymbol{r},t)$ [10].

The yielding of a site leads to a rearrangement with a local deformation rate given by

$$\partial_t \epsilon^{pl}(\boldsymbol{r},t) = n(\boldsymbol{r},t)\varepsilon(\boldsymbol{r},t)/(2\tau)$$

τ=1 mechanical relaxation time, $n(\boldsymbol{r},t)$ local state variable, n=1:yielding , n=0:not-yielding.

$\varepsilon(\boldsymbol{r},t) = \pm\varepsilon_0$ depending on the yielding direction. Two symmetric thresholds $\sigma_Y = \pm\sigma_0$.

Response of the surrounding medium modeled by Eshelby theory of elasticity with the elastic kernel G(**r**,**r'**). Over-damped dynamics for the coarse-grained scalar stress field

$$\partial_t \sigma(\boldsymbol{r},t) = 2\mu \int d\boldsymbol{r}' G(\boldsymbol{r},\boldsymbol{r}') \partial_t \epsilon^{pl}(\boldsymbol{r}';t)$$

μ elastic modulus, $G^\infty(r,\theta) = 2\cos(4\theta)/\pi r^2$. Discretized time and space (square lattice $L \sim 2^8, 2^9$, periodic boundary conditions). Dynamics solved using a pseudo-spectral method.

Rules for $n(\boldsymbol{r},t)$ are stochastic; sites with $|\sigma| > |\sigma_Y|$ become immediately active (n:0→1), while for sites with $|\sigma|<|\sigma_Y|$:

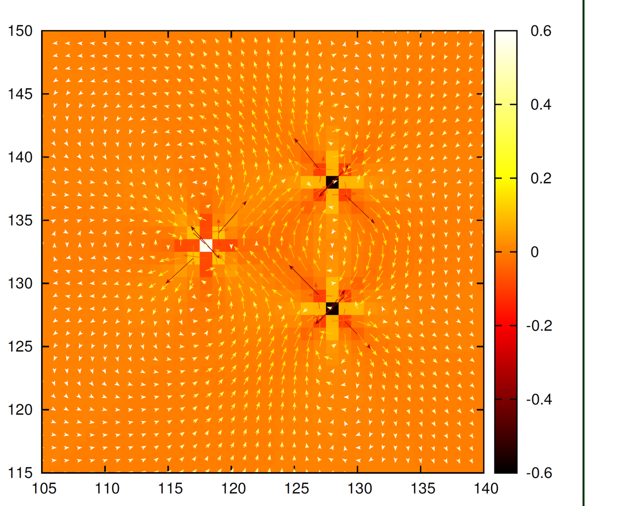
$$p_{\mathtt{on}}(\pm) = \Gamma_0 \exp\left[\frac{-(\sigma_Y^2 \mp \mathtt{sgn}(\sigma)\sigma^2)}{2\kappa T}\right]$$

Active sites deactivate (n:1→0) at a fix rate

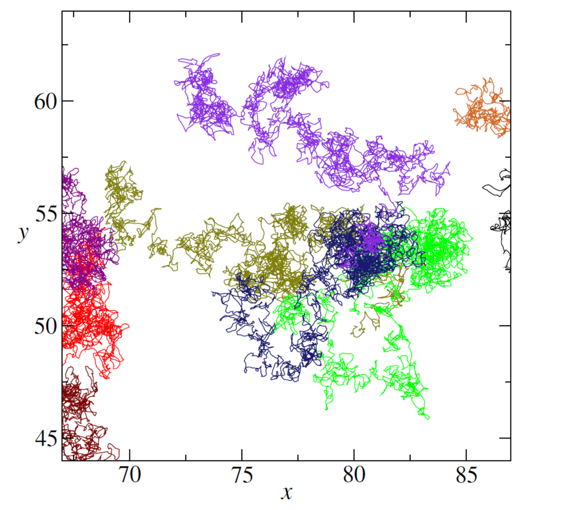
$$p_{\mathtt{off}} = \tau_{\mathtt{res}}^{-1}$$

Γ_0=1 attempt frequency, τ_{res}=1 typical duration of a restructuring event, $\kappa = \mu V_0^{-1} k_B$ provide correct magnitudes. For each temperature T, a steady state is reached (stress fluctuations around zero with a Gaussian distribution of stresses).

At each time step the vectorial displacement field $\boldsymbol{u}(\boldsymbol{r},t)$ associated with the discretized plastic strain field is calculated [10]. Tracer particles follow this field, with no further interactions. Form their trajectories we build the quantities of interest.



Displacement fields for three simultaneous plastic events



Typical tracers trajectories in the (x,y) plane

Auxiliary *random* model

Analogous system with the sole difference of having randomly activated sites, instead of a stress-dependent rule. We measure the mean activity a(the number of events per unit time) in the thermal model and plug it in the random model as a controlled parameter to compare equivalent systems.

$$a = \left\langle \frac{1}{N} \sum_i n_i \right\rangle$$

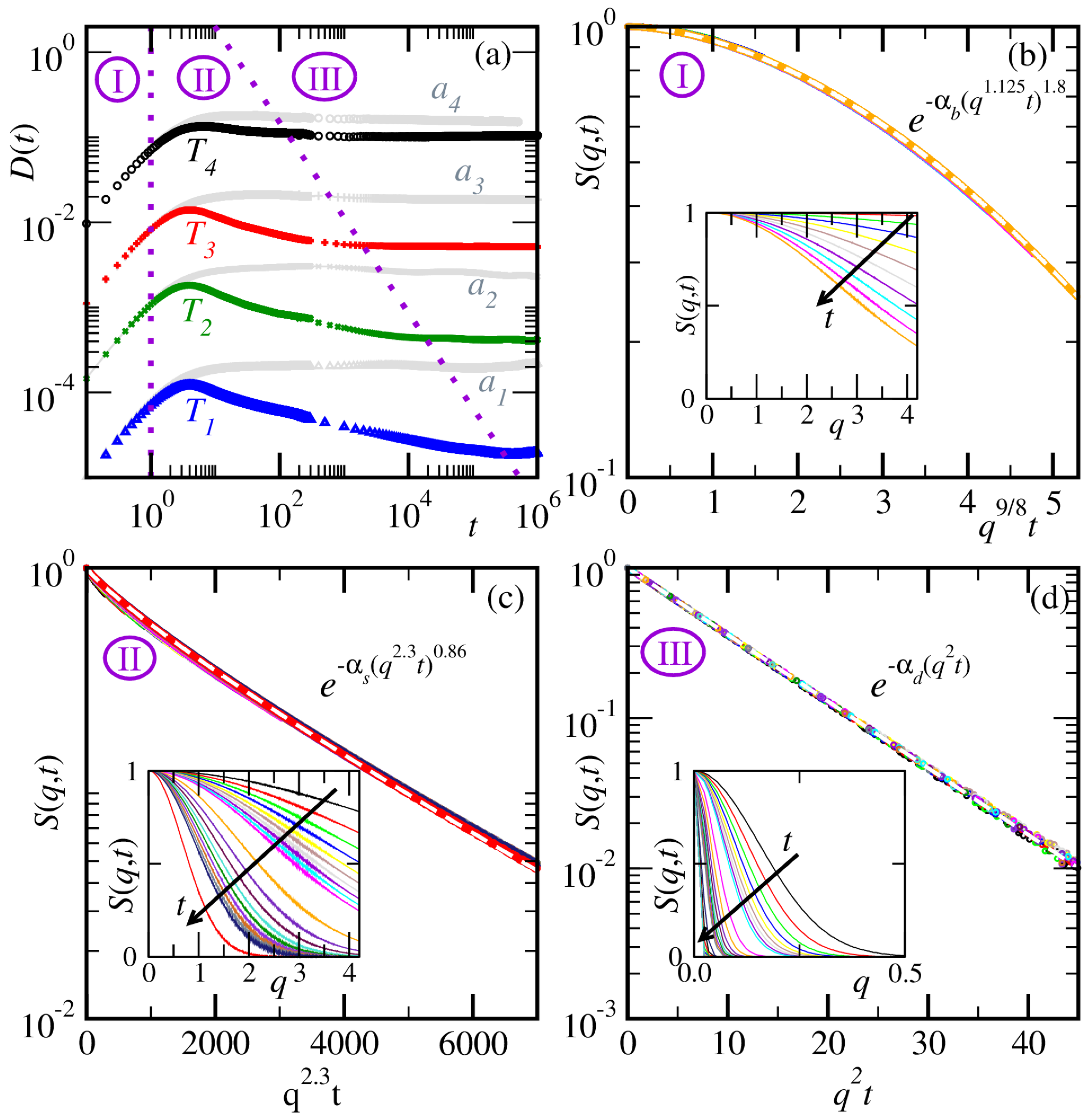


Fig.1: ***Tracer particle dynamics*– (a)** Diffusion coefficient $D=(\Delta r)^2/(4t)$ as a function of time t for different temperatures T_i = 0.05, 0.07, 0.1, 0.2. Three dynamical regions: (I) ballistic, (II) crossover, (III) diffusive. Gray curves show results for the random model with a mean activity $a_i \approx 3.1e^{-0.48/T_i}$. **(b)** Dynamical structure factor $S(q,t)$ for T=0.2 as a function of $q^{1.125}t$ for time intervals corresponding to the ballistic regime (I), fitted by an compressed exponential with shape parameter $\gamma \approx 1.8$ and $\alpha_b \approx 0.07$ (dashed line). **(c)** $S(q,t)$ for T=0.07 as a function of $q^{2.3}t$ for the crossover regime (II), fitted by a stretched exponential with $\gamma \approx 0.86$ and $\alpha_s \approx 0.0015$ (dashed line). **(d)** $S(q,t)$ for T=0.2 as a function of q^2t for the diffusive regime (III), fitted by a pure exponential with $\alpha_D \approx 0.1$ (dashed line). Insets in (b,c,d) show raw data for each case.

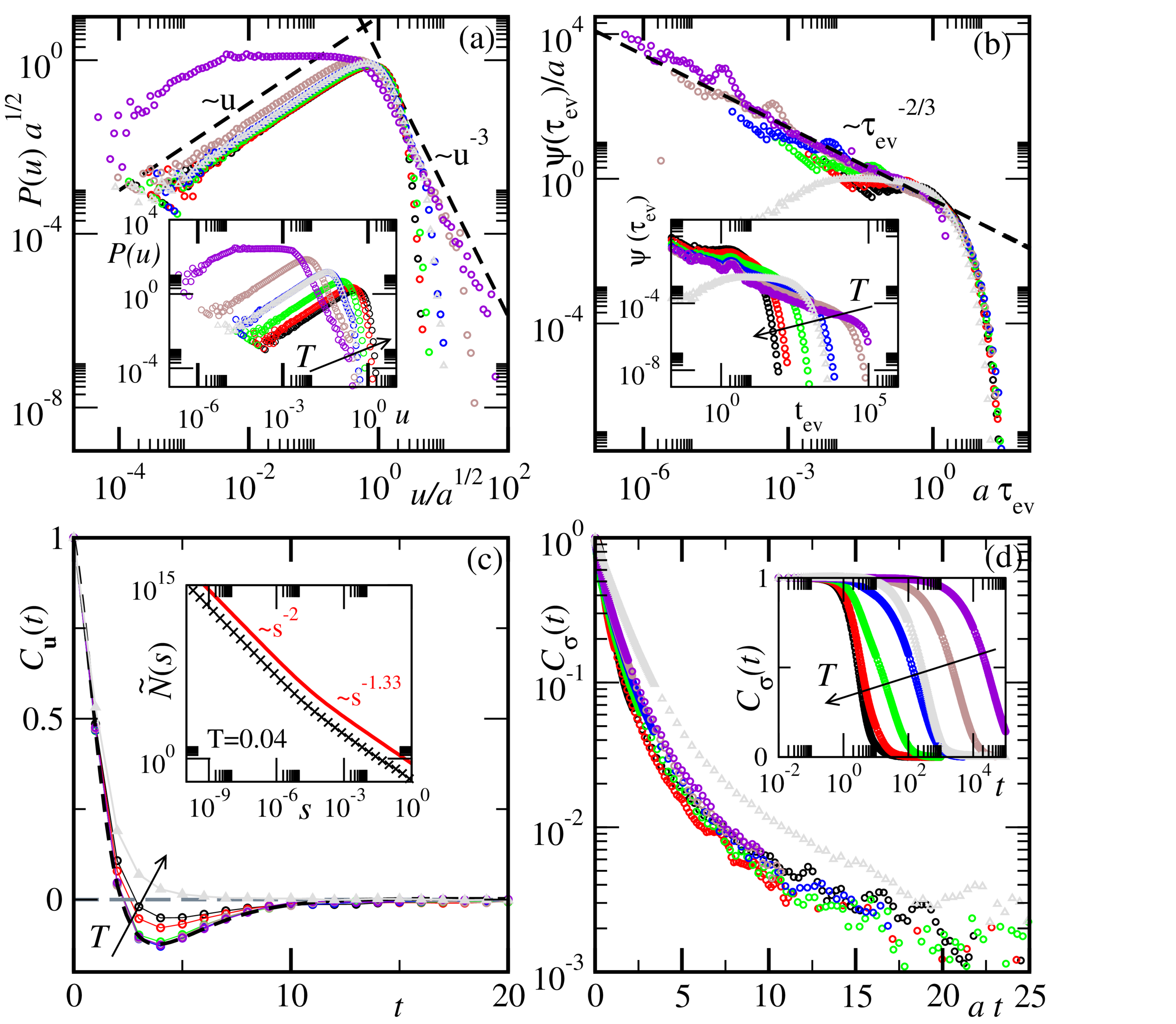


Fig.2: ***Statistical features of the thermally activated dynamics*** – In all panels, circles correspond to the thermal model for temperatures T_i = 0.04, 0.05, 0.07, 0.1, 0.15, 0.2, while gray triangles stand for the random model at a≈0.0034. **(a)** Distribution of absolute displacements per unit time $u=|\boldsymbol{u}|$ of the tracer particles for different temperatures, rescaled by the square-root of the average activity $a(T_i)$. Dashed lines display power-laws. **(b)** Rescaled local probability distribution $\psi(\tau_{ev})/a(T_i)$ for rescaled elapsed times $a(T_i)\tau_{ev}$ between events. The power-law dashed line is as a guide to the eye. **(c)** Normalized two time auto-correlation function $C_{\boldsymbol{u}}(t)$ of the vectorial displacements of tracer particles. Inset: result of a CTRW model with the Laplace transform of $\psi(a\tau_{ev})$ as input. **(d)** Normalized two-time auto-correlation function $C_\sigma(t)$ of the local stress as a function of the rescaled time $a(T_i)t$. Insets in (a,b,d) show data without rescaling.

Results

Elasticity effects

- At each temperature we observe for $D(t)$: an **initial ballistic regime** independent of the model, of duration ruled by τ_{res}, and a **long time diffusive** behavior (Fig.1a)
- We fit $S(q,t)$ in the **ballistic** regime with a **compressed exponential** of shape parameter close to expected MF value in 2d γ_{MF}=2 (Fig.1b), in the **diffusive** regime we obtain a **pure exponential** decay on q^2t, as expected (Fig.1d).
- Regarding the distribution of particle **displacements** (independent on the model) we confirm the expected **mean-field scalings** for low temperatures (Fig.2a).

Correlation effects

- In the **thermal model**, we observe for $D(t)$ an **intermediate sub-diffusive regime** with a duration ruled by T (Fig.1a). During this regime, the relaxation time τ_r scales as $\tau_r \sim q^{-n}$ with n>2. Relaxation curves collapse onto a **stretched exponential** master curve $S(q,t) = \exp[\alpha_s(q^n t)^\gamma]$ with γ<1 (Fig.1c).
- Since the distribution $P(u)$ is identical in both models, the change of the dynamics for the **thermal model** is rather due to **negative correlations** in the two-time auto-correlation function of the **vectorial displacements** (Fig.2c).
- The **distribution of waiting times** $\Psi(\tau_{ev})$ (trivially exponential in the random model) shows a **power-law** form with an exponential cutoff in the thermal model (Fig.2b). Plugging in $\Psi(\tau_{ev})$ in **a simplified CTRW** model the mean square displacement already **shows the crossover** between sub-diffusive an diffusive behavior (Inset Fig.2c).

Discussion

- **Compressed exponential** relaxation can result from an **elastic response** of the medium to **thermally activated plastic events**, in a time scale comparable with their duration. The commonly referenced mean-field compressed shape parameter γ_{MF}=3/2 is valid in 3d only, in 2d γ_{MF}=2.
- Even when **aging** is typically present in all experimental studies reporting compressed exponentials, and actually affects the typical relaxation time, **it is not necessarily a key ingredient** to observe the compressed relaxation.
- **Beyond** the **mean field** results we find that **correlations** between events **generate a partial confinement** of the tracers. Instead of enhancing the persistence of the tracer particles as often assumed in the literature, correlations lead in our model to sub-diffusive behavior.

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