

Soft Matter

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Spatial correlations of elementary relaxation events in glass-forming liquids

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Received Xth XXXXXXXXXXXX 20XX, Accepted Xth XXXXXXXXXXXX 20XX

First published on the web Xth XXXXXXXXXXXX 200X

DOI: 10.1039/b000000x

The dynamical facilitation scenario, by which localized relaxation events promote nearby relaxation events in an avalanching process, has been suggested as the key mechanism connecting the microscopic and the macroscopic dynamics of structural glasses. Here we investigate the statistical features of this process via the numerical simulation of a model structural glass. First we show that the relaxation dynamics of the system occurs through particle jumps that are irreversible, and that cannot be decomposed in smaller irreversible events. Then we show that each jump does actually trigger an avalanche. The characteristic of this avalanche change on cooling, suggesting that the relaxation dynamics crosses over from a noise dominated regime where jumps do not trigger other relaxation events, to a regime dominated by the facilitation process, where a jump trigger more relaxation events.

1 Introduction

Structural glasses, which are amorphous solids obtained by cooling liquids below their melting temperature avoiding crystallization, provide an array of questions that has been challenging researchers in the last decades^{1–3}. These include the nature of the glass transition, the origin of the extraordinarily sensitivity of the relaxation time on temperature, the Boson-peak, the relaxation dynamics. In this respect, here we consider that there is not yet an established connection between the short time single particle motion, and the overall macroscopic dynamics. When observed at the scale of a single particle, the motion of structural glasses is well known to be intermittent. This is commonly rationalized considering each particle to rattle in the cage formed by its neighbors, until it jumps to a different cage⁴. Conversely, when the motion is observed at the macroscale, a spatio-temporal correlated dynamics emerges⁵. Dynamical facilitation^{6–8}, by which a local relaxation event facilitates the occurrence of relaxation events in its proximity, has been suggested as a key mechanism connecting the microscopic and the macroscopic dynamics. Indeed, kinetically constrained lattice model⁹, which provide the conceptual framework of the dynamical facilitation sce-

nario, reproduce much of the glassy phenomenology and are at the basis of a purely dynamical interpretation of the glass transition. Different numerical approaches have tried to identify irreversible relaxation events^{10–20}, and both numerical^{21,22} and experimental works^{23,24} revealed signatures of a dynamical facilitation scenario.

Here we provide novel insights into the dynamical facilitation mechanisms through the numerical investigation of a model glass former. We show that it is possible to identify single particle jumps that are *elementary* relaxations, being short-lasting irreversible events that cannot be decomposed in a sequence of smaller irreversible events. We then clarify that these jumps lead to spatio-temporal correlations as each jump triggers subsequent jumps in an avalanching process. The statistical features of the avalanches changes on cooling. Around the temperature where the Stokes–Einstein relation first breaks down, the dynamics shows a crossover from a high temperature regime, in which the avalanches do not spread and the dynamics is dominated by thermal noise, to a low temperature regime, where the avalanches percolate. These results suggest to interpret dynamical facilitation as a spreading process²⁵, and might open the way to the developing of dynamical probabilistic models to describe the relaxation of glass formers.

2 Methods

We have performed NVT molecular dynamics simulations²⁶ of a two-dimensional 50:50 binary mixture of $2N = 10^3$ of

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disks, with a diameter ratio $\sigma_L/\sigma_S = 1.4$, known to inhibit crystallization, at a fixed area fraction $\phi = 1$ in a box of side L . Particles interact via a soft potential²⁷, $V(r_{ij}) = \varepsilon((\sigma_{ij} - r_{ij})/\sigma_L)^\alpha \Theta(\sigma_{ij} - r_{ij})$, with $\alpha = 2$ (Harmonic). Here r_{ij} is the interparticle separation and σ_{ij} the average diameter of the interacting particles. This interaction and its variants (characterized by different values of α) are largely used to model dense colloidal systems, such as foams²⁸, microgels²⁹ and glasses^{30,31}. Units are reduced so that $\sigma_L = m = \varepsilon = k_B = 1$, where m is the mass of both particle species and k_B the Boltzmann's constant. The two species behave in a qualitatively analogous way, and all data presented here refer to the smallest component.

Cage-jump detection algorithm. We segment the trajectory of each particle in a series of cages interrupted by jumps using the algorithm of Ref.³², following earlier approaches¹¹. Briefly, we consider that, on a timescale δ of few particle collisions, the fluctuation $S^2(t)$ of a caged particle position is of the order of the Debye–Waller factor (DWF) $\langle u^2 \rangle$. By comparing $S^2(t)$ with $\langle u^2 \rangle$ we therefore consider a particle as caged if $S^2(t) < \langle u^2 \rangle$, and as jumping otherwise. Practically, we compute $S^2(t)$ as $\langle (r(t) - \langle r(t) \rangle_\delta)^2 \rangle_\delta$, where the averages are computed in the time interval $[t - \delta : t + \delta]$, with $\delta \simeq 10t_b$, and t_b is the ballistic time. At each temperature DWF is defined according to Ref.³³, $\langle u^2 \rangle = \langle r^2(t_{DW}) \rangle$, where t_{DW} is the time of minimal diffusivity of the system, i.e. the time at which the derivative of $\log \langle r^2(t) \rangle$ with respect to $\log(t)$ is minimal. At each instant the algorithm allows to identify the jumping particles and the caged ones. We stress that in this approach a jump is a process with a finite duration. Indeed, by monitoring when S^2 equals $\langle u^2 \rangle$, we are able to identify the time at which each jump (or cage) starts and ends. We thus have access to the time, t_p , a particle persists in its cage before making the first after an arbitrary chosen $t = 0$ (persistence time), to the waiting time between subsequent jump of the same particle t_w (cage duration), and to the duration Δt_j and the length Δr_j of each jump.

3 Results

3.1 Jumps as irreversible elementary processes

The idea of describing the relaxation of structural glasses as consisting of a sequence of irreversible processes is not new, and different approaches have been followed to identify these events. For instance, irreversible events have been associated to change of neighbors^{13–15}, to displacements overcoming a threshold in a fixed time laps²², to processes identified through clustering algorithm applied to the particle trajectories^{21,23}, or to more sophisticated approaches¹⁷. We notice that since at long time particles move diffusively, all procedures that coarse grains the particle trajectory enough will eventually identify ir-

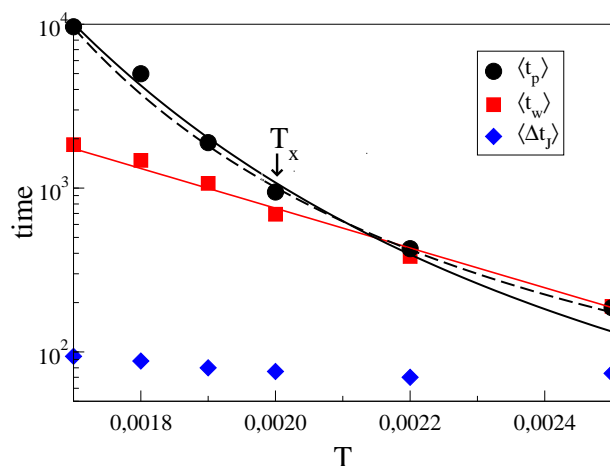


Fig. 1 Average persistence time, $\langle t_p \rangle$, cage duration, $\langle t_w \rangle$ and jump duration, $\langle \Delta t_j \rangle$, as a function of the temperature. $\langle t_w \rangle$ grows as an Arrhenius $\langle t_w \rangle \propto \exp(A/T)$ (red full line), whereas $\langle t_p \rangle$ is compatible with several super-Arrhenius laws. The black full line is, for example, a fit $\langle t_p \rangle \propto \exp(A/T^2)$, while the black dashed line is a Vogel–Fulcher law $\langle t_p \rangle \propto \exp(B/(T - T_0))$, predicting a divergence at a finite temperature $T_0 \simeq 0.001$. The arrow indicates the temperature $T_x = 0.002$ where $\langle t_p \rangle$ and $\langle t_w \rangle$ decouple and the SE relation breaks down. Conversely, $\langle \Delta t_j \rangle$ remains roughly constant on cooling.

reversible events. Here we show that the jumps we have identified are irreversible, and we give evidence suggesting that these can be considered as ‘elementary’ irreversible events, i.e. that they are the smallest irreversible single-particle move, at least in the range of parameters we have investigated.

Investigating both the model considered here³², as well as the 3d Kob-Andersen Lennard-Jones (3d KA LJ) binary mixture³⁴ and experimental colloidal glass³⁵, we have previously shown that the protocol defined in Sec. 2 leads to the identification of irreversible events. Indeed, the mean square displacement of the particles increases linearly with the number of jumps, allowing to describe the dynamics as a continuous time random walk (CTRW)³⁶.

Within this approach two fundamental timescales are found, the average persistence time $\langle t_p \rangle$ and the average cage duration $\langle t_w \rangle$. The former corresponds to the relaxation time at the wavelength of the order of the jump length $\langle \Delta r_j \rangle$, while the latter is related to the self diffusion constant, $D \propto \langle \Delta r_j^2 \rangle / \langle t_w \rangle$. Fig.1 shows that the two timescales are equal at high temperature, but decouple at a temperature $T_x \simeq 0.002$, which marks the onset of the Stokes-Einstein (SE) breakdown at the wavelength of the jump length. We find that $\langle t_w \rangle$ shows an Arrhenius temperature dependence $\langle t_w \rangle \propto \exp(A/T)$, while $\langle t_p \rangle$ increases with a faster super-Arrhenius behaviour (see the caption of Fig.1). It is worth noticing that the decoupling between

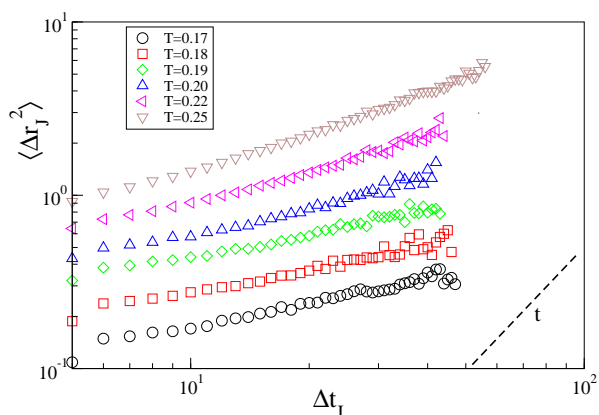


Fig. 2 Mean squared jump length $\langle \Delta r_J^2 \rangle$ as a function of the jump duration Δt_J at different temperatures.

the average persistence and waiting time, is known to control the breakdown of the SE relation at generic wavelengths, and to induce temporal heterogeneities^{34,37}. These findings suggest that T_x may represent a crossover from a localized to a more correlated relaxation process. A similar scenario has been recently reported for models of atomic glass forming liquids, where the SE breaks down and the size of dynamics heterogeneities markedly accelerates below a well defined value of T_x .³⁸

We performed two investigations supporting the elementary nature of the jumps we have identified. First, we have considered the change of the average jump duration $\langle \Delta t_J \rangle$ on cooling, as the duration of elementary relaxations is expected not to grow with the relaxation time. Fig. 1 shows that the $\langle \Delta t_J \rangle$ is essentially constant, despite the relaxation time $\langle t_p \rangle$ varying by order of magnitudes. Indeed, at low temperature $\langle t_p \rangle / \langle \Delta t_J \rangle \gg 1$, clarifying why we call them ‘jumps’. Then we have considered how particles move while making a jump. Fig. 2 illustrates that the mean squared jump length grows subdiffusively as a function of the jump duration, with a subdiffusive exponent that decreases on cooling. Conversely, one would expect a diffusive behaviour if jumps were decomposable in a series of irreversible steps.

These results supports the identification of the jumps we have defined with the elementary relaxations leading to the macroscopic relaxation of the particle system.

3.2 Correlations between jumps

While each particle behaves as a random walker as it performs subsequent jumps, yet jumps of different particles could be spatially and temporally correlated. We investigate these correlations focusing on the properties of a jump birth scalar field,

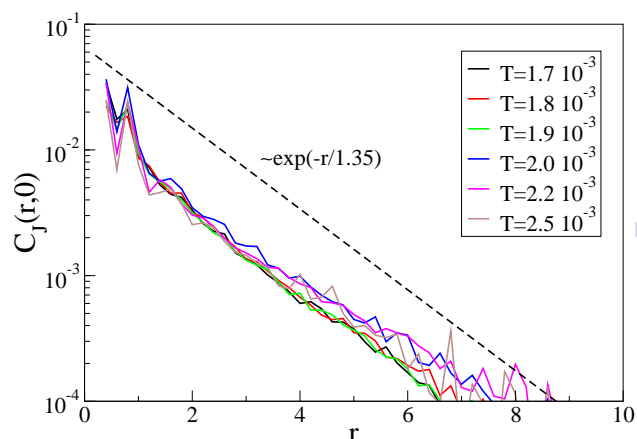


Fig. 3 Excess probability to observe contemporary jumps, $C_J(r,0)$, as function of the distance and at different temperature, as indicated. The dashed line is a guide to the eyes $\propto \exp(-1.35/r)$.

defined as

$$b(r,t) = \frac{1}{N} \sum_i^N b_i(t) \delta(r - r_i(t)). \quad (1)$$

Here $b_i(t) = 1$ if particle i starts a jump between t and $t + \delta t$, where δt is our temporal resolution, $b_i(t) = 0$ otherwise. The scalar field b allows to investigate the statistical features of the facilitation process by which a jump triggers subsequent ones. To this end, we indicate with $\langle b(r,t) \rangle_{b(0,0)=1}$ the probability that a jump starts in (t,r) given a jump in $(t=0, r=0)$, and investigate the correlation function

$$C_J(r,t) = \left[\frac{\langle b(r,t) \rangle_{b(0,0)=1} - \langle b \rangle}{g(r,t)} \right]. \quad (2)$$

Here $g(r,t)$ is a time dependent generalization of the radial distribution function

$$g(r,t) dr = \frac{1}{2\pi r \rho (N-1)} \sum_{i \neq j} \delta(r - |r_j(t) - r_i(0)|), \quad (3)$$

through which we avoid the appearance of spurious oscillations in the correlation function $C_J(r,t)$ due to the short range ordering of the system. In Eq.2, $\langle b \rangle$ is the spatio-temporal average of the jump birth, and decreases on cooling as $\langle b \rangle = (\langle t_w \rangle + \langle \Delta t_J \rangle)^{-1}$ (at low temperature $\langle b \rangle \simeq \langle t_w \rangle^{-1}$ as $\langle t_w \rangle \ll \langle \Delta t_J \rangle$). Accordingly, the correlation function $C_J(r,t)$ is the probability that a jump triggers a subsequent one at distance r after a time t .

We first consider the spatial correlations between contemporary jumps, where two jumps are considered contemporary if occurring within our temporal resolution. Fig. 3 shows that $C_J(r,0)$ decays exponentially, with a temperature independent correlation length $\xi_J(0,T) \simeq 1.35$. This result clarifies that

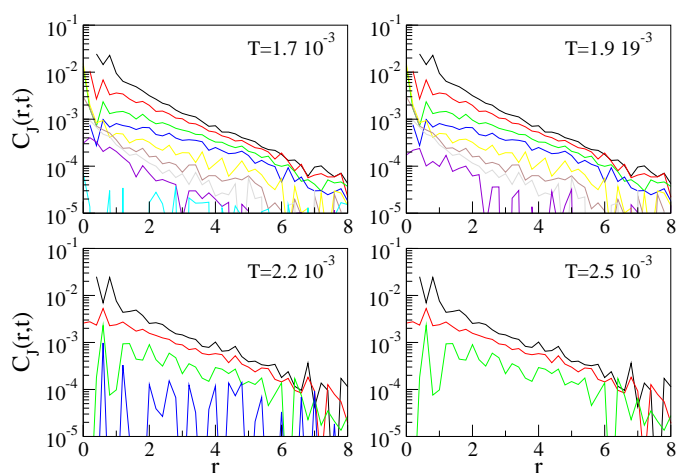


Fig. 4 Evolution of the spatial correlation between jumps with time. Each panel refer to a different temperature, as indicated. Within each panel, the different curves correspond to $t = 0, 10, 20, 30, 100, 500, 10^3$ and 10^5 , from top to bottom. At high temperature data corresponding to the largest times are missing as the correlation is too small to be measured.

jumps aggregate in cluster of roughly $N_{corr} \simeq \rho \pi \xi_J^2(0) \simeq 5$ events. A similar scenario has been observed in a different model system, where jumps have been observed to aggregate in clusters of roughly 7.6 particles²¹. Our results also support previous findings suggesting²² that the elementary excitations of structural glasses have a temperature-independent length not larger than few particle diameters and are consistent with a recently introduced first principle extension of the Mode Coupling Theory³⁹. The investigation of the displacements of the particle jumping in each cluster does not reveal characteristic spatial features. Structured particle motion, such as string-like particle displacements⁴⁰ or displacements reminiscent of T1 events⁴¹ must therefore result from a succession of events rather than a single one.

We now consider the time evolution of the spatial correlation between jumps. Fig. 4 illustrates that at all temperatures and times the decay of the correlation function is compatible with an exponential, $C_J(r,t) \propto A(t) \exp(-r/\xi_J(t))$. The time dependence of the amplitude is illustrated in Fig. 5. At all temperatures the short time decay of the amplitude is exponential, $A(t, T) = A(0, T) \exp(-t/\tau_A(T))$, the characteristic decay time slightly increasing on cooling. While no other decay is observed at high temperatures, at low temperatures the exponential decay crossovers towards a much slower power-law decay $A(t) \sim t^{-a}$, with $a \simeq 0.4$. Fig. 6 shows that the correlation length slowly grows in time, approximately as $\xi_J(t) \sim t^b$, with $b \simeq 0.1$.

The initial fast decrease of the amplitude makes difficult to

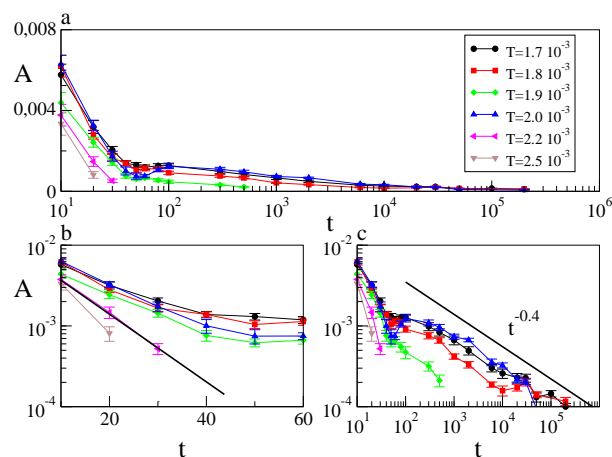


Fig. 5 Panel a shows that the amplitude $A(t)$ of the jump correlation function $C_J(r,t)$. Panels b and c clarify that a first exponential decay is followed, at low temperature, by a second one, which approximately follows a power law.

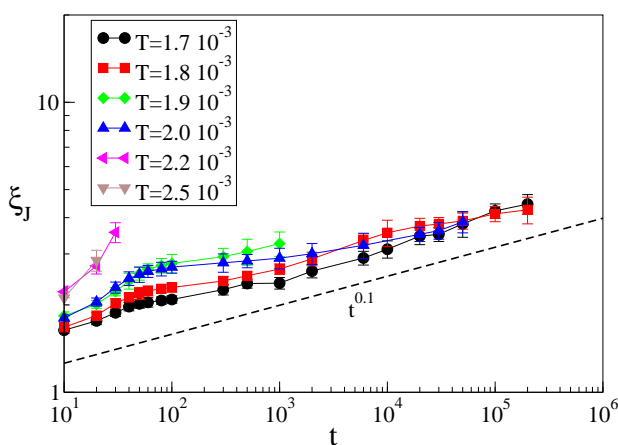


Fig. 6 Time dependence of the jump correlation length, at different temperatures. The data suggest that at low temperature the correlation length slowly grows in time, as $\xi_J(t) \propto t^{0.1}$.

obtain reliable estimates of its time dependence and correlation length, despite intense computational efforts. Nevertheless, our data clearly show the reported exponential to power-law crossover in the decay of the amplitude of $C_J(r,t)$. The highest temperature at which this decay exhibits a power law tail, is consistent with the temperature T_x where $\langle t_w \rangle$ and $\langle t_f \rangle$ first decouple, and the SE relation breaks down (see Sec.3.1). This suggests that the breakdown of the SE relation is related to a crossover in the features of the facilitation process. We investigate this crossover focussing on the number of jumps triggered by a given jump. This is given by $N_{tr}(T) \propto \int_0^\infty n(t, T) dt$, where $n(t, T) = \int C(\mathbf{r}, t) \mathbf{r} d\mathbf{r} \propto A(t, T) \xi_J^2(t, T)$, is the num-

ber of jumps it triggers at time t . As at high temperature the variation of the correlation length is small with respect to that of the amplitude, one can assume $\xi(t, T) \simeq \xi(0, T)$ and estimate $N_{\text{tr}}(T) \propto A(0, T)\xi^2(0, T)\tau_A(T)$. At low temperature, the integral is dominated by the long time power law behavior of the amplitude and of the correlation length, and the number of triggered events diverges as $N_{\text{tr}}(T, t) \propto \int_0^t A(t)\xi^2(t)dt \propto t^{-a+2b+1} \propto t^{0.8}$.

4 Discussion

We conclude by noticing that the above scenario suggests to interpret facilitation as an infection spreading process, in which a particle is infected each time it jumps. Since each particle can be infected more than once, the relevant infection model is of susceptible-infected-susceptible (SIS) type. In this framework, the exponential to power-law crossover in the decay of the amplitude of $C_J(r, t)$ signals a transition from a high temperature resilient regime, in which a single infected site only triggers a finite number of infections, to a low temperature regime in which the number of triggered infection diverges. A complementary interpretation can be inspired by the diffusing defect paradigm^{1,42}. We suggest that the correlation length of contemporary jumps, $\xi_J(0)$, is akin to the typical defect size, which, according to our results, is temperature independent. In the high temperature regime, this is the only relevant correlation length, as defects are rapidly created and destroyed by noisy random fluctuations, before they can sensibly diffuse. At low temperature, the effect of noise becomes smaller: the short time correlation length is still dominated by the defect size, $\xi_J(t < \tau_A) \simeq \xi_J(0)$, whereas its long time behaviour, $\xi_J(t \gg \tau_A)$, is controlled by the typical distance defects have moved up to time t . Further studies are necessary to investigate which of the two interpretations is more appropriate.

Acknowledgement

We acknowledge financial support from MIUR-FIRB RBF081IUK, from the SPIN SEED 2014 project *Charge separation and charge transport in hybrid solar cells*, and from the CNR-NTU joint laboratory *Amorphous materials for energy harvesting applications*.

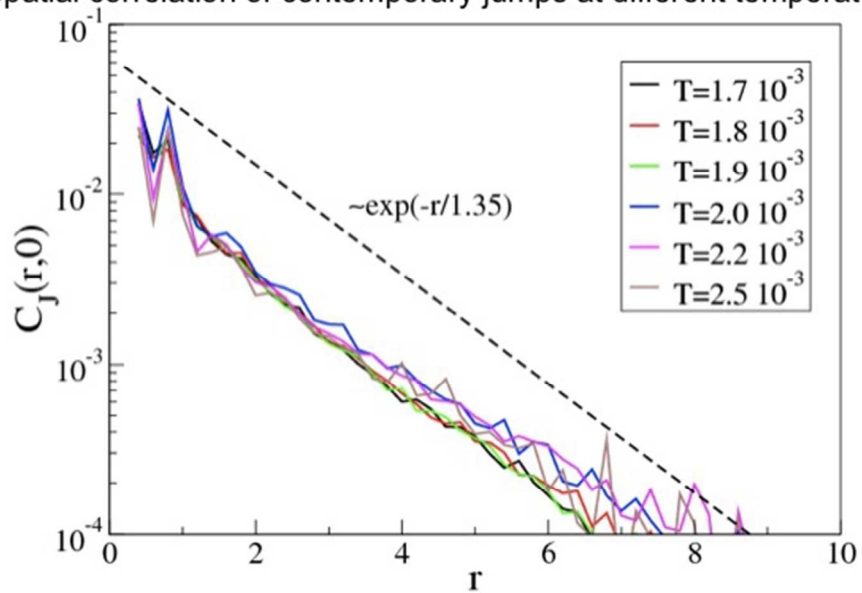
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We identify the smallest irreversible events in glass formers and investigate their correlations.

Spatial correlation of contemporary jumps at different temperature



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