Crackling Noise during Slow Relaxations in Crumpled Sheets

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Crackling noise during slow relaxations in crumpled sheets

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The statistics of noise emitted by ultra-thin crumpled sheets is measured while they exhibit logarithmic relaxations under load. We find that the logarithmic relaxation advanced via a series of discrete, audible, micro-mechanical events that are log-Poisson distributed (i.e., the process becomes a Poisson process when time stamps are replaced by their logarithms). The analysis places constraints on the possible mechanisms underlying the glass-like slow relaxation and memory retention in these systems.

Introduction- Studying the characteristics of noise emitted by a dynamic system has long been used as a tool to gain insights into the system's properties and ongoing physical processes [1]. For instance, careful measurements of shot noise in electronic devices has been used as an indicator for fractional charge and exotic phases in certain electronic systems [2], and the study of $1/f$ noise in structural glasses has similarly led to fruitful insights regarding the nature of the two-level-systems postulated to be present in this system [3].

Here we use noise measurements to gain insight into the mechanisms underlying slow relaxation, aging and memory effects, which are some of the hallmark behaviors of out-of-equilibrium disordered systems. These behaviours are exhibited by a wide range of systems, such as structural and electron glass [4], frictional interfaces [5] and colloidal systems [6], as well as disordered mechanical systems such as granular piles [7]. Slow mechanical relaxations under constant load are also referred to as "creep" [8], including strain relaxation under a constant load of ice [9], metals [10], rock [11] as well as the silk threads used by Weber in his pioneering experiments on magnetism [12]. We also note that in Ref. [13] we have shown that analogous behavior is obtained in a protocol where stress relaxation is measured for a fixed strain.

One system that exhibits these behaviours is a large, thin sheet of Mylar that has been crumpled many times into a ball [14]. When placed under constant uniaxial load, the system's volume exhibits an ever-slowing relaxation process that spans many time scales - from fractions of a second to weeks, without showing any signs of reaching equilibrium. This behavior is reproduced in Fig. 1a, as explained below. Similar phenomena is observed in the normal force exerted by a crumpled thin sheet when it is placed under constant strain [13, 15]. Previously, we have shown that when this system is subjected to a two-step protocol in which after a controlled waiting time, t_w, the applied load or strain is abruptly changed to another fixed value, the ensuing relaxation, while still slow, exhibits clear *non-monotonic* features. In particular, under constant conditions, a macroscopic observable (volume or normal force) increases slowly over times scales ranging from seconds to hours, comes to a halt and then decreases, converging to a logarithmic relaxation [13]. This non-monotonic memory dynamics is similar to the celebrated Kovacs effect that has been observed in a range of glassy systems such as polymer melts [16, 17], metallic glasses [18] and granular systems [19].

Here, we analyze the acoustic emissions emitted by crumpled thin sheets during logarithmic volume relaxation under load, and use the results to constrain the possible mechanisms leading to slow relaxation and memory retention in this system. While here we focused on the logarithmic compaction of crumpled sheets under constant load, as mentioned the same system was shown to exhibit *memory effects* during relaxation in response to load perturbations [13]. Earlier experiments also noted that crumpled thin sheets emit crackling noise when strained [20, 21]. However, so far these measurements were performed only during manual manipulation and were focused on the statistics of the intensities of the acoustic emissions. In this work we show that the crackling patterns measured during logarithmic relaxation of crumpled sheets under load are consistent with a stochastic version of the model introduced in Ref. [13] to explain relaxation and memory in these systems.

Experiments- A thin sheet of Mylar, 500X500 millimeters in size and 12 micrometers in thickness was crumpled for more than 50 times into a ball and then placed under a load of 200 grams. The height of the system was measured using a magnetic displacement sensor, sampled at 22KHz using a data acquisition card for over six hours. The displacement data was averaged offline to a rate of 5Hz, to reduce the measurement noise. The results indicate a nearly perfect logarithmic relaxation spanning more than four decades in time, as shown in Fig. 1a. As the system relaxes under the load, it emits audible crackling sounds, which is recorded using an amplified microphone and a 1KHz high-pass filter in a noise-isolated chamber, at a rate of 22KHz (the spectra of the crack-

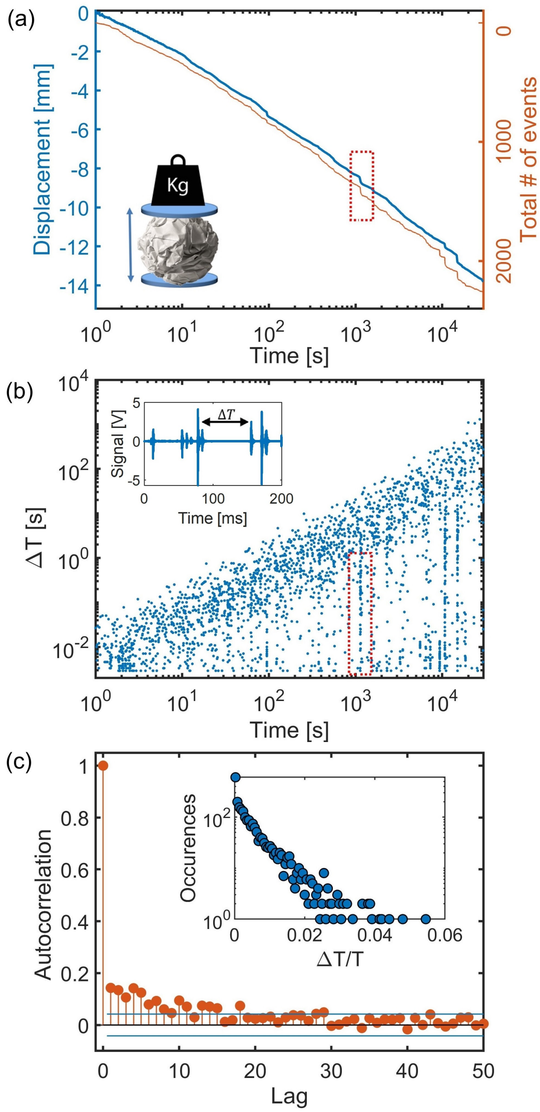

FIG. 1. Acoustic emission during logarithmic relaxation of crumpled sheets (a) Measurements of the height of a ball of crumpled thin sheet under load as a function of time, showing logarithmic relaxation (blue line) and the accumulated number of acoustic events during the relaxation (orange). (b) log-log plot of the time gaps between consecutive acoustic emissions, on the same time axis as the experiment. inset: example of the measured acoustic signal. Note the appearance of avalanches (red dotted rectangle) that correlated with intermittent features in the relaxation curve. (c) Measured auto-correlation of time gaps between consecutive acoustic emission, normalized by the time passed since the beginning of the experiment. Inset: histogram of normalized time gaps, showing approximate log-Poisson statistics.

ling sounds emitted by the crumpled sheets in the experiments are typically centered around 2KHz). An example of an acoustic recording is attached.

A small sample of the obtained acoustic emission signal is shown at the inset of Fig. 1b, revealing a series of discrete pulses. Each pulse has a typical duration of about 2 milliseconds, and a central frequency of about 2000Hz, and are easily detected in the large raw data files. During a typical experiment we detect more than 2000 acoustic emission events, as shown in Fig. 1.

Fig. 1b shows the time gaps between each two consecutive acoustic emission events. Three important observation can be made from this plot. First, the average time gap between consecutive events increases linearly with the age of the experiment: while in the first seconds of the experiment the typical time gap is about 10 milliseconds, this number increases to about 500 seconds by the end of the experiment. Second, most of the events are concentrated in a narrow band which retains a width of about one decade throughout the experiment. Third, occasionally many events happen during a short time, with time gaps between consecutive events that quickly drops to the lower temporal detection limit. One such avalanche is marked by a red dotted rectangle in Fig. 1b. The accumulated number of acoustic events correspondence clearly to the height relaxation, from the slight curvature of the plot down to small details, as shown in Fig 1a. In particular, the avalanches detected in the acoustic emission measurements manifest in a sharp steps in the height relaxation curve, as highlighted for the same avalanche in Fig. 1a and 1b.

The statistics of time gaps between consecutive acoustic emission events is shown in Fig 1c. The inset shows a histogram of the time gaps, each divided by the time since the beginning of the experiment, to normalize out the linear slowing-down of the process. The result is an approximate Poisson distribution of the normalized time gaps, indicative of an approximate log-Poisson statistics for the time gaps themselves. The auto-correlation function for the sequence of normalized time gaps indicates a weak correlation that reduced to noise after about 20 events, as shown in Fig. 1c. This short-time correlation, as well as the first point in the histogram, are mainly a result of the rare avalanches.

The experimental observations indicate that the mechanical relaxation of the crumpled sheet is accompanied by a sequence of discrete, audible, micro-mechanical events, suggesting that each event is releasing some of the internal stress and contributing to the overall displacement of the system. The sensitivity of height measurement in our logarithmic relaxation experiment is not sufficient to detect the height decrease due to any single event. However, the comparison between the curves, Averaged over 15 repetitions of the experiment performed with the same sheet yields an average displacement 5.5 ± 0.8 microns per acoustic event. The robust ratio between the number of events and the displacement (either across measurement or due to an avalanche) is indicative that this sequence of micro-mechanical events are indeed related to the process by which the system relaxes.

Continuous model- Our starting point is a model we introduced previously to account for logarithmic relaxation and Kovacs-like memory response in crumpled sheets [13]. We have shown, experimentally, that when subjected to a constant (uniaxial) strain, the force exerted by a crumpled thin sheet decays logarithmically over many decades in time, in line with earlier measurements on a similar protocol where a constant force is applied to the sheet and the strain changes logarithmically [14]. Moreover, we used a two-step protocol to demonstrate that the system exhibits clear aging behavior: the applied strain was E_1 for a duration t_w and then switched to a different strain E_2, and the resulting slow relaxations depended explicitly on both t_w and the measurement time t. The function $F(t_w, t)$ provides important information regarding the physical mechanisms underlying slow relaxations. Intriguingly, despite the complexity of this system we found that the function F showed non-monotonic relaxations with a peak at time t_p scaling *linearly* with t_w.

This simple result was consistent with a model in which the relaxations are a superposition of many exponential relaxations, drawn from the distribution of relaxation rates $P(\lambda) \propto 1/\lambda$ (with lower and upper cutoffs). Moreover, this formalism captured the logarithmic form of the relaxations. In fact, this distribution was derived originally within a mean-field model for slow relaxations in electron glasses. There, linearization of the equations governing the system dynamics led to [22] $\frac{d\vec{v}}{dt} = A\vec{v}$ with $\vec{v}$ a high-dimensional vector corresponding to all degrees of freedom of the system, and A a matrix whose (negative) eigenvalues correspond to the relaxation rates, and whose spectrum was later shown to follow the aforementioned $1/\lambda$ distribution [23]. This model was used to predict the aging dynamics in electron glasses [24]. Note that within this interpretation, the amplitude associated with each eigenmode decays exponentially – and smoothly.

Adopting this model for the crumpled sheets, this model captures the non-monotonic relaxations and quantitatively explains the linear scaling between t_p and t_w and the logarithmic relaxations, and thus seems to adequately describe the dynamics of the system. Within this mean-field approximation and to the broad distribution of eigenvalues, if one furthermore assumes that the external perturbation is sufficiently small such that the eigenmodes (and eigenvalues) are nearly identical in the presence and absence of the perturbation, one finds a particular form of aging of the form:

$$f(t, t_w) \propto \log(t + t_w) + C \log(t), \tag{1}$$

where the constant C depends on the ratio of the external fields E_1 and E_2. When a field is switched on for a time t_w and then switched off, $C = -1$, and f collapses to the form of "full" or "simple" aging $f(t, t_w) \propto \log(1 + t_w/t)$, which has been demonstrated to hold for several glasses [8, 25]. Here, the *reversibility* of the modes played an important role: the amplitude corresponding to each mode increases when the perturbation is applied and thereafter relaxes exponentially.

Nevertheless, the interpretation of the slow dynamics of the crumpled sheets as arising from a superposition of exponentially relaxing eigenmodes of the linearized dynamics is incompatible with the experimental observation described above, showing that the relaxation advances via a series of discrete, audible events. Next, we suggest a simple model to reconcile the measured aging behavior with the discrete nature of the noise. Moreover, the model leads to approximately log-Poisson statistics of the discrete relaxation events, which appears to be consistent with the experiments. Finally, within the revised model we will not demand reversibility of the relaxation modes – the effective description using the above model will arise statistically, without invoking reversibility of individual modes.

A stochastic model.- As in the phenomenological theory of relaxations in structural glasses, we consider an ensemble of two-level-systems (TLS), each characterized by a barrier U between the two states and a bias Δ corresponding to the energy difference between the two states. As TLS theory, we assume that the energy U is uniformly distributed within some interval of width W. We assume that the rate of transition from the higher energy state to the lower energy state is given by $\lambda = e^{-U/T_{eff}}$, where $T_{eff} \ll W$ is a constant (which in thermally-driven systems corresponds to the temperature). For the relaxations in the crumpled sheets we assume that the transitions are irreversible. We also assume that the application of a force affects the value of Δ, such that a TLS which was previously in its lowest state may be "destablized". Such a TLS will ultimately return to its ground state, albeit via a stochastic process whose rate per unit time is λ. The transition is assumed to lead to a strain relaxation by a small but finite amount accompanied by a discernible "click". SM.IV provides a visual comparison of simulations of this model, in contrast to the continuous one.

We comment that the memory and aging behavior within this model are *identical* to the one discussed above. If we consider all TLS with the same λ, then after time t_w the fraction of those which remain in the higher energy metastable state is $e^{-\lambda t_w}$ – hence they will play the same role as a single exponentially decaying eigenmode of that relaxation rate. Nevertheless, note that within this model the dynamics is stochastic and it is only upon averaging over all modes with the same λ that one recovers the aforementioned continuous picture.

Next, we proceed to find that statistics of the discrete relaxation events generated by this model, in the one-step protocol (in which a stress is applied to the system and maintained). Note that in aforementioned aging experiments rather than measuring the strain for a given stress the opposite is done – the strain is fixed and the stress relaxation quantified. We do not expect this to qualita-

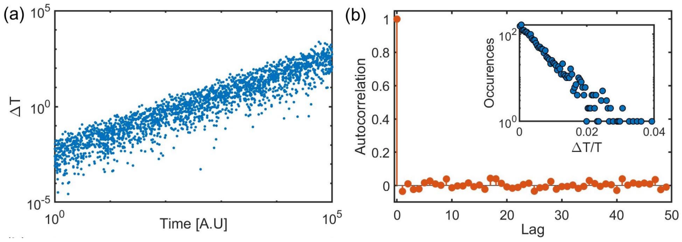

FIG. 2. Stochastic simulation of the crackling statistics assuming a broad relaxation rate distribution (3000 modes with rates between 10^{-6} and 10). (a) The typical time gaps between consecutive events increases linearly with time, as predicted theoretically. (b) Since the model assumes transitions between *independent* two-level-systems, the autocorrelation of time gaps vanishes for nonzero lags. Inset: Upon rescaling time, the time gap distribution becomes Poisson, as shown analytically in the main text.

tively change any of the conclusions, and base our discussion on the former protocol, following Ref. [14], where this striking phenomenon was first observed. *Comparing Theoretical and Experimental Results.-* Consider the system at time t. The probability that an unstable mode with relaxation rate λ has not yet relaxed is $e^{-\lambda t}$. Since each of these are independent and memoryless, the distribution of times to the next relaxation event is Poisson, with a rate:

$$\Lambda = \sum_j \lambda_j = \int_0^\infty e^{-\lambda t} \lambda P(\lambda) d\lambda. \quad (2)$$

In our case $P(\lambda) \propto 1/\lambda$, and we find: $\Lambda = \int_{\lambda_{min}}^{\lambda_{max}} e^{-\lambda t} d\lambda$, where $\lambda_{min,max}$ are the cutoffs of the relaxation rate distribution. Under the assumption that the experimental timescales are far from these cutoffs, we find $\Lambda \propto 1/t$. This implies that at time t the mean time to the next relaxation event is proportional to the time t, and exponentially distributed:

$$P(\Delta t) = e^{-A\Delta t/t}. \quad (3)$$

Moreover, the proportionality constant A will be inversely proportional to the system size, since the number of TLS must be extensive.

Eq. (3) describes the slowing down of the system: as time goes by the time between relaxation events increases. If we rescale Δt by the measurement time t, we find that it will follow Poisson statistics. This is very similar (but not identical) to the log-Poisson statistics discussed in the context of noise in other glassy systems [26]: For sufficiently large systems we have $\langle \Delta t \rangle = 1/\Lambda \ll t$, in which case we may approximate:

$$x_{n+1} - x_n = \log(t + \Delta t) - \log(t) \approx \Delta t/t, \quad (4)$$

where x_i is the logarithm of the time of the i'th click. Since $\Delta t/t$ is Poisson distributed, we conclude that the logarithm of event times approximately follows Poisson statistics. This is tested numerically in Fig. 2b (inset).

In the SM.I we also derive the click statistics without invoking the approximation used in Eq. (3), distinct from the predictions of the record statistics model of Ref. [26].

Noise in the aging regime.- In the previous derivation, the slowing down of the clicking rate scaled as $1/t$, which is intuitive since the relaxation is logarithmic. If each click corresponds to a release of a constant amount of energy, then the rate of change of the potential energy is compatible with the click statistics. One may therefore naively expect that the time derivative of the height to determine the click statistics.

This intuition is in fact incorrect. To appreciate the problem, it is instructive to consider the *two-step* protocol discussed above, which in previous works we and others have found to provide more information and constrain the possible model space. In the case $E_1 < E_3 < E_2$, the magnitude of the coefficient C of Eq. (1) is smaller than 1, and the relaxation curve is non-monotonic – implying that the sign of the signal derivative flips. Therefore clearly the noise rate cannot be proportional to it.

In SM.II we calculate the noise in the aging regime, which makes for a clear theoretical prediction; One obtains a Poisson distribution for click statistics, $p(\Delta T) \propto e^{-r\Delta t}$ with parameter $r(t)$:

$$r(t) = \frac{2E_3 - E_1 - E_2}{t + t_w} + \frac{E_2 - E_3}{t}. \quad (5)$$

At long times $t \gg t_w$ one recovers the $(E_3 - E_1)/t$ scaling – as expected. Note that when the non-monotonic relaxations reach a peak, the click statistics are not expected to vanish – at this point the two subpopulations have an identical switching rate. These predictions are corroboration in the SM.III, where we find good agreement between theory and experiments.

Discussion.- Here, we have studied, theoretically and experimentally, the statistics of noise in a crumpled, thin sheet undergoing slow relaxations of displacement under constant load. By modifying the model previously proposed for electron glasses, we reconcile the discrete nature of the relaxation events manifested by short acoustic emissions, with the logarithmic relaxations observed over many decades in time: the statistics are interpreted to arise due to stochastic transitions between metastable states, resulting in a macroscopic description mathematically equivalent to that of an ensemble of *reversible* two-level-systems – a reversibility which played a key role in explaining the aging behavior in this system in previous work, and in particular the emergence of non-monotonic relaxations. We also predict the noise statistics in this aging regime, yet to be tested experimentally. Note that within this study the system is always out-of-equilibrium: this is distinct from previous studies showing that glasses with logarithmic, slow relaxations will also exhibit $1/f$ noise in their fluctuations around a metastable state [27].

In fact, a variety of models were suggested over they years to account for logarithmic relaxation in disordered systems, including renewal processes [28] and record statistics [26]. The latter also predicts the log-Poisson statistics discussed here and also observed experimentally in colloidal glasses[29]. However, these mechanisms do not quantitatively explain the particular aging results reported previously for crumpled thin sheets [13]. Interestingly, there are parallels between our results here and those observed for creep of rocks under uniaxial stress, which also exhibit logarithmic time-dependence of strain. Ref. [11] interpreted this logarithmic relaxation by considering failure of local regions, occurring with a rate that is Arrhenius (i.e. exponentially) dependent on the local parameters, assumed to be inhomogeneous. This bears resemblance to the phenomenological model we propose here, albeit with the distinction that while in brittle rocks each region can fail once and in an irreversible manner, the results of Ref. [13] illustrate the for the crumpled sheets each "two-level-system" must be able to respond in a reversible manner – this behavior lies at the heart of the Kovacs-like, non-monotonic relaxations. The analogy with brittle rock goes further, as, remarkably, Ref. [30] was able to record the acoustic emissions associated with the logarithmic creep, using high-precision accelerometers. Their findings are parallel to the results of the crumpled sheet, finding that acoustic emissions are localized in time, and that the rate of these events falls off as $1/t$.

Future studies should determine the microscopic origin of the phenomenological model discussed here – do the TLS correspond to bistable mechanical configurations, recently shown to underlie the mechanics of crumpled sheets [20, 31]? If so, what are their spatial extents, and how do they mechanically interact with each other? Note that within the current model, interactions were not included. Furthermore, it is intriguing to find whether these phenomena are related to recent work showing the evolution of crease-formation in crumpled, thin sheets, where a logarithmic dependence of the total crease length with the number of crumpling events was shown both experimentally [32] and theoretically [33].

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