

## Memory in Nonmonotonic Stress Relaxation of a Granular System

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(Received 4 November 2019; revised manuscript received 21 February 2020; accepted 30 March 2020; published 20 April 2020)

We demonstrate experimentally that a granular packing of glass spheres is capable of storing memory of multiple strain states in the dynamic process of stress relaxation. Modeling the system as a noninteracting population of relaxing elements, we find that the functional form of the predicted relaxation requires a quantitative correction which grows in severity with each additional memory and is suggestive of interactions between elements. Our findings have implications for the broad class of soft matter systems that display memory and anomalous relaxation.

DOI: [10.1103/PhysRevLett.124.168002](https://doi.org/10.1103/PhysRevLett.124.168002)

Subjected to a perturbation, many systems in nature will relax anomalously (i.e., nonexponentially) over long timescales, suggesting complex dynamics and common underlying, far-from-equilibrium physics. Examples of specifically logarithmic relaxation are the slow breaching of colloidal particles at an interface [1], the magnetization decay in type-II superconductors [2], the dynamics of crumpled elastic sheets [3,4], the evolving area of frictional contact between two interfaces [5,6], and the stress decays in a granular packing at fixed strain [7,8].

A versatile framework applied to the relaxation of viscoelastic [9] and dielectric materials [10–12] idealizes a system as an ensemble of simple, exponential relaxers in parallel with one another, with a distribution of different relaxation times (DRT). To explain the widespread occurrence of logarithmic relaxation, Amir *et al.* [13] motivated a specific distribution of relaxation times on fairly general grounds. Importantly, the Amir, Oreg, and Imry (AOI) variant of DRT was also used to explain two-step non-monotonic relaxation observed in crumpled mylar [4], a frictional interface [6], and bulk rock salt [14], after subjecting such systems to a particular driving protocol.

Nonmonotonic relaxation is a surprising and nonintuitive phenomenon. In the process of energy dissipation, with no external input after the initial driving, a state variable evolves in one direction before turning around after some timescale that was imprinted during the prior driving history. In contrast with memories that are revealed only when the system is driven [15], these memories reside in dynamic processes and thus offer a foothold into studying the quixotic march to equilibrium of a far-from-equilibrium system.

Here we employ a granular packing to study nonmonotonic relaxation within the DRT framework. We store the memory of an additional strain state in experiment by appending a compression step at the end of a two-step driving protocol. The functional form of the relaxation, while qualitatively similar to that predicted by the model, requires a quantitative correction which grows in severity

with the additional memory. We suggest a route to reconciliation between the model and experiment, guided by the presence of discrete relaxation events in the data.

For each experiment, 5 mm diameter soda lime glass spheres (MoSci) were poured into a 5 cm diameter latex membrane to form a 2:1 column (height to diameter) [Fig. 1(a)]. Isotropic confinement of 40 kPa was applied via holding the interior of the membrane at lower pressure. The column was compressed by an Instron universal materials tester to a strain of  $\epsilon = 0.1$ , as a fraction of the initial (uncompressed) height, over 2 min. During this initial compression, stress builds across the packing and grain reconfigurations lead to stress fluctuations [Fig. 1(b)]. All experiments in this Letter began in this way and then were followed by up to two additional strain steps  $\Delta\epsilon^{(i)}$  after wait times  $t_w^{(i)}$ . The stress relaxation of interest occurs in the final static hold, during which nothing further is done to the granular system beyond monitoring the stress.

Simple uniaxial compression followed by a hold without any additional steps of (de)compression leads to stress relaxation of the granular material that is approximately logarithmic in time [Fig. 1(c)], in agreement with past results [7,8]. That the relaxation is nonexponential suggests granular materials might be able to exhibit nonmonotonic relaxation after an appropriate driving protocol, and the logarithmic form in specific invites a treatment with the AOI distribution of relaxation rates, as in Refs. [4,6].

One possible protocol to initiate nonmonotonic relaxation is as follows: rather than holding the system at a strain state  $\epsilon$  indefinitely, it is allowed to relax partially for some time  $t_w^{(1)}$ , but then the applied strain is *decreased* to  $\epsilon - \Delta\epsilon^{(1)}$ . After driving a granular packing in this way, it relaxes in a strikingly nonintuitive manner: without any additional prompting, nor any additional energy input to the system, the stress measured *increases* for a period of time before turning around and resuming a slow decrease that shows no signs of stopping on experimental timescales.

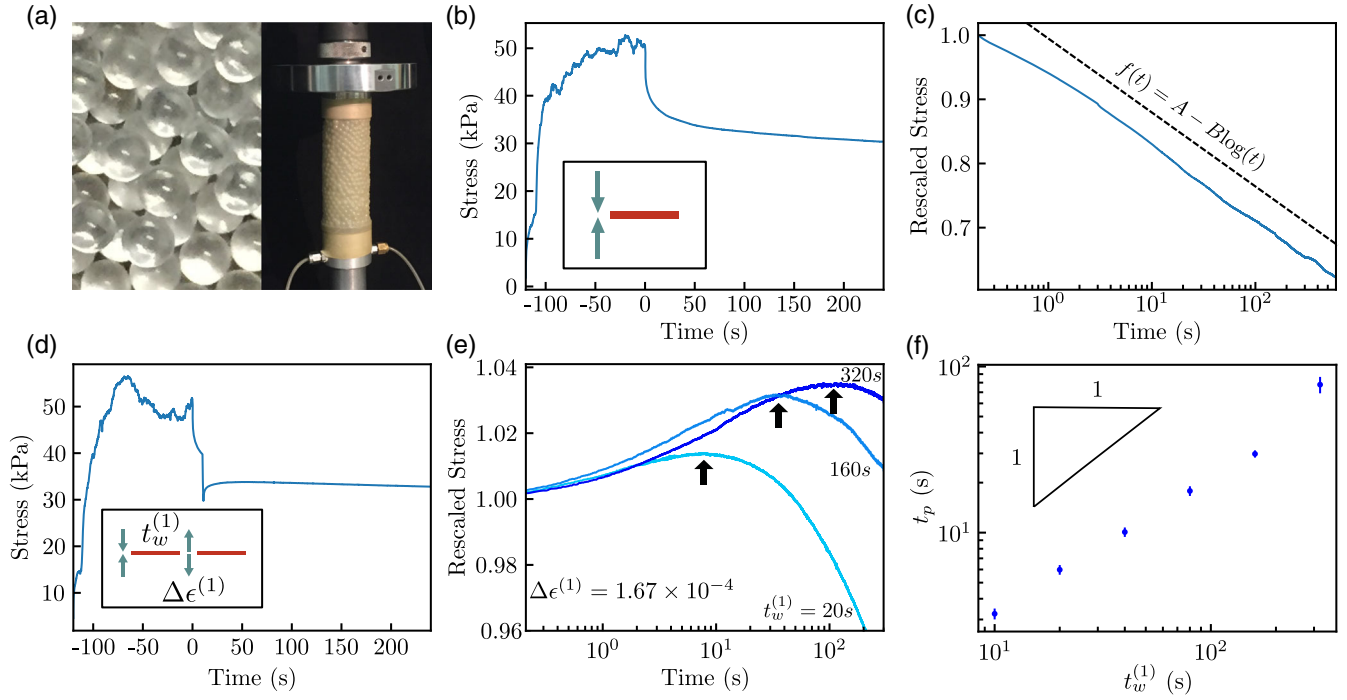


FIG. 1. Anomalous relaxation in a granular packing of glass spheres. (a) The 5 mm glass spheres used in experiment, magnified and in the latex membrane. (b) Stress-time data for single step relaxation, where a packing is compressed to strain  $\epsilon = 0.1$  and then held statically for the remainder of the experiment. Inset illustrates the experimental protocol: arrows pointing inward (outward) correspond to compression (decompression), and red lines denote holds at constant strain. (c) The stress decays approximately logarithmically from the moment compression halts. These data are from (b), with stress normalized by its value at the start of the hold. (d) Stress-time data for two-step relaxation. (e) The stress of two-step relaxation for three different  $t_w^{(1)}$ , with turnaround time marked by arrows. (f) The turnaround time  $t_p$  scales approximately linearly with  $t_w^{(1)}$ , for constant  $\Delta\epsilon^{(1)}$ .

Further, the timescale  $t_w^{(1)}$  of the hold in the strain state  $\epsilon$  emerges as a memory that is revealed in the turnaround time  $t_p$  [Figs. 1(d), 1(e)].

We employ DRT to explain the nonmonotonic relaxation, as was previously done by Lahini *et al.* [4]. Within the framework of DRT, a system is idealized as a population of simple, exponential relaxers in parallel with a distribution of relaxation rates  $P(\lambda)$ . In the AOI variant [13],  $P(\lambda) \sim 1/\lambda$  over a range  $[\lambda_{\min}, \lambda_{\max}]$ , yielding logarithmic relaxation over timescales between  $\lambda_{\max}^{-1}$  and  $\lambda_{\min}^{-1}$ . Compelling reasons in support of this distribution of relaxation rates were described in earlier work on luminescence [16].  $P(\lambda) \sim 1/\lambda$  is uniform in  $\log\lambda$  space and identical whether working in terms of rates  $\lambda$  or timescales  $\tau = \lambda^{-1}$ . Both  $\lambda$  and  $\tau$  are scale parameters—i.e., domain of  $(0, \infty)$ —and as such this distribution is the maximum entropy distribution [17] for which minimal prior information has been assumed. The distribution maximizes generality, providing a reason for the widespread occurrence of logarithmic relaxation.

Each of the relaxing elements holds a portion of stress  $\Gamma(\lambda)$  which dissipates exponentially in time according to  $d\Gamma = -\lambda\Gamma dt$ , and the measured signal (i.e., the total stress) is the sum over all elements. Compression affects all elements equally and is taken to occur over a timescale

negligible to even the fastest elements [i.e., a (de)compression step takes  $\Gamma(\lambda) \leftarrow \Gamma(\lambda) + \alpha\Delta\epsilon$ , with  $\alpha$  a constant]. Figure 2(c) shows the system state at various stages of relaxation, where the elements are displayed from slowest (left, dark) to fastest (right, light). At a time  $t_w$  into the relaxation [Fig. 2(c) II], the fastest elements have relaxed to  $\Gamma = 0$  stress and the slowest still bear most of their original stress.

In such a state, the system has dual natures: through the slow elements it remembers the initial, unstrained state, and through the fast ones it has adapted to the strain state  $\epsilon$ . Decompression at this time decreases the magnitude of the stress in the slow elements and negatively stresses the fast elements, creating a system state  $\Gamma(\lambda)$  in which subsets of the elements will relax in opposing directions. The fast elements relax first, causing the paradoxical increase in stress over time even though all elements decrease in energy, which scales with the square of the stress. Eventually the slow elements turn the relaxation around, giving rise to nonmonotonic dynamics. The memory is clearly visible in the stress held across the relaxing elements,  $\Gamma(\lambda)$ , in state III of Fig. 2(c), where the timescale of the switch in sign was imprinted by the duration of the hold  $t_w^{(1)}$ .

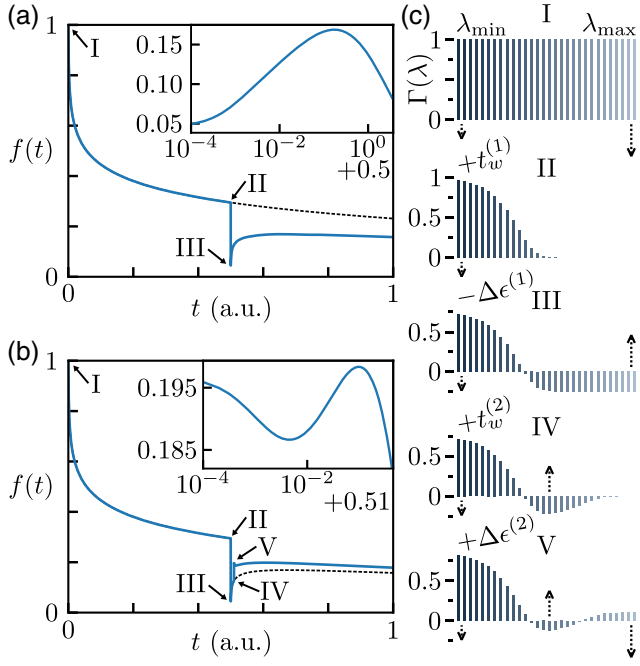


FIG. 2. The different relaxation times framework (DRT) with the Amir-Oreg-Imry distribution of relaxation rates (AOI). (a) Simulated stress-time data for two-step relaxation. Inset: The relaxation from the start of the final hold. (b) Simulated data for three-step relaxation. Inset: The relaxation from the start of the final hold. In (a) and (b) the dashed line shows the relaxation of  $f(t)$  without the extra strain step  $\Delta\epsilon^{(1)}$  and  $\Delta\epsilon^{(2)}$ , respectively. (c) The state of the system  $\Gamma(\lambda)$  at various points in time, shown as the stress held by each element ordered from slowest (dark, left) to fastest (light, right). The time of the states are marked in (a) and (b) as Roman numerals I–V. The arrows show the direction and qualitative magnitude of relaxation. The scale of  $\Gamma(\lambda)$  is relative to the stress in each element in state I.

Casting the nonmonotonic relaxation of the granular packing within the DRT framework shows that the storage capacity of memories should be larger than 2. Specifically, by appending a small strain step in the original (positive) direction, we can create three steps of relaxation in a simulated system [Fig. 2(b)]. Again, the memory is manifest in  $\Gamma(\lambda)$  of state V in Fig. 2(c), where the population of relaxing elements has been split into three counterrelaxing contingents.

Guided by the simulated system, we find three-step relaxation—the first observed in any disordered system, to the best of our knowledge—in our packings by adding a small compression step  $\Delta\epsilon^{(2)}$  in the forward direction, after waiting time  $t_w^{(1)}$  at  $\epsilon$  and then  $t_w^{(2)}$  at  $\epsilon - \Delta\epsilon^{(1)}$  [Fig. 3(a)]. The resulting stress during relaxation undulates back and forth without any intervention: it decreases, increases, and then decreases again over timescales imprinted during the loading process. After fixing the strain state for the final hold, nothing is done to the granular system to prompt the nonmonotonicity; thus at the start of the hold the packing is

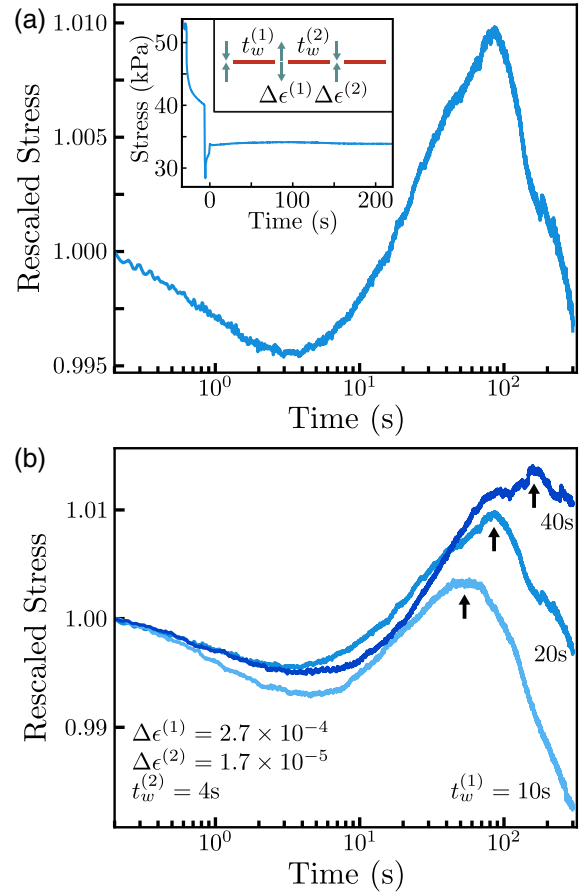


FIG. 3. Three-step relaxation of glass beads. (a) Stress during the final hold of three-step relaxation, rescaled by its value at the start. Inset: Stress-time data for the various stages of relaxation, showing the two wait times  $t_w^{(1)}$  and  $t_w^{(2)}$  before the final hold. (b) Varying  $t_w^{(1)}$  while holding the rest of the driving protocol constant shifts the time of the second turnaround in stress. Memories are thus played out in the reverse order from which they were stored during driving.

in a state that “knows” to turn around in stress after 4 sec and then again some 90 sec later.

The DRT model with the AOI distribution yields a functional form for the nonmonotonic relaxation: a sum of alternating logarithms, staggered in time according to the start of the prior strain steps [4] [see Figs. 4(a), 4(b)]. However, we find a correction is necessary to achieve reasonable fits to the two- and three-step relaxation data (Fig. 4). Specifically, we find that the wait times inside the logarithms need to be multiplied by a parameter  $C > 1$ . This does not arise from the model (where  $C = 1$ ) and indicates deficiency in its descriptive power. Importantly, only a significant modification to the model could scale the waiting timescales  $t_w^{(i)}$  in this way; changes to  $P(\lambda)$  or the excitation upon compression  $\Delta$  do not rescale  $t_w^{(i)}$  [18].  $C > 1$  was also necessary to fit the two-step relaxation in Ref. [4], though no attention was called to it, presumably

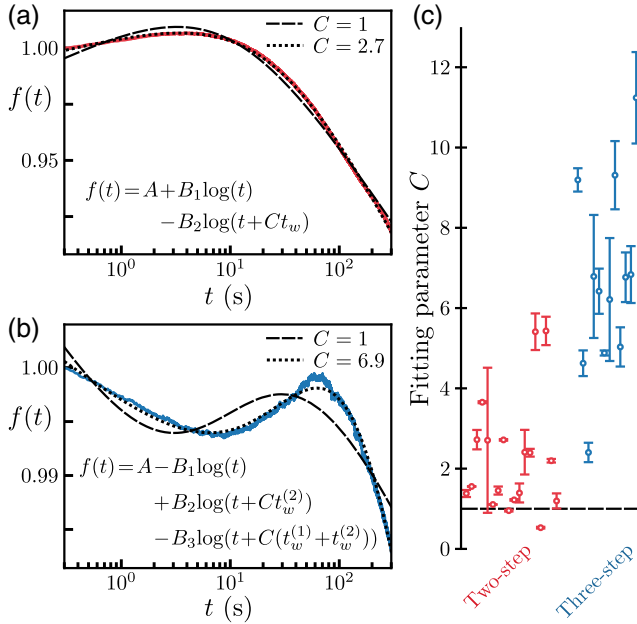


FIG. 4. Fitting multistep relaxation data to the form predicted by the model. (a) An example of two-step relaxation data fitted to the form predicted by the model ( $C = 1$ ) and to a form where  $C$  is allowed to vary. (b) The same, for an example of three-step relaxation. (c) The best-fit values of the parameter  $C$  across all two- and three-step relaxation experiments. The dashed line,  $C = 1$ , represents DRT with the AOI distribution.

because  $C$  was close to unity. We find that three-step relaxation data significantly increases the discrepancy between the AOI DRT prediction and the experimental results, with fitted  $C$  values often an order of magnitude larger than the model allows [Fig. 4(c)].

We conjecture about  $C > 1$  based on observations of discrete events that occur during the relaxation of a granular packing. During such events, the stress or its derivative suddenly change in magnitude. These events appear to be the same local restructuring events that constitute plastic deformation under continued compression [Fig. 1(b), and [26]]. In many cases, one of which is shown in Fig. 5(a), an event renews fast timescale relaxation late into a static hold. In terms of the DRT framework, the event effectively “rejuvenated” faster relaxing elements which had long since adapted to the current strain state. This is suggestive of crosstalk between the relaxing elements, occurring in discrete steps during events such as the one shown and ostensibly continuously during the gradual flow of the granular packing. That the renewed relaxation is approximately logarithmic over more than 2 orders of magnitude in time shows the crosstalk redistributed stress to the fast elements nearly uniformly.

A simple form of crosstalk between elements can be incorporated into the model through an effective diffusion of stress in  $\Gamma(\lambda)$ . The diffusion is included as a Laplacian in  $\log \lambda$  space, scaled by a coefficient  $D$ . Implemented in this way, all relaxation curves [Fig. 5(b)] are fit by the same

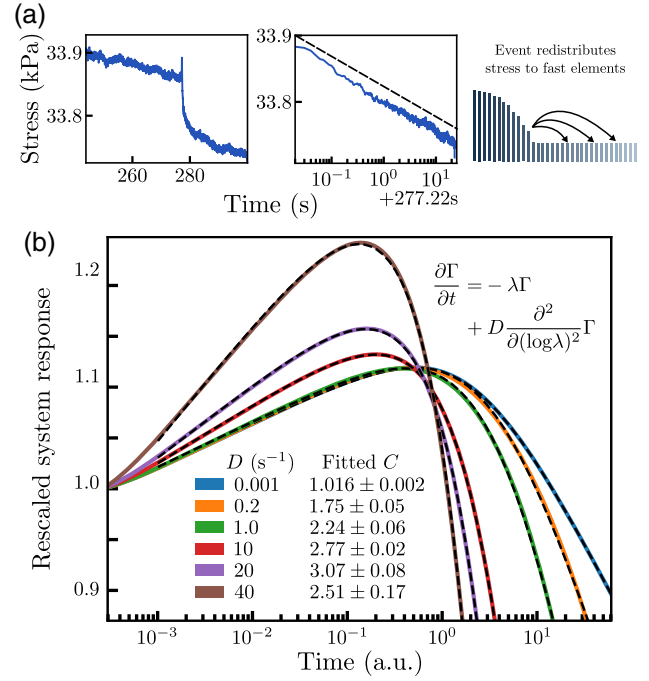


FIG. 5. Crosstalk between elements in the model. (a) A sudden relaxation event late into one of the experiments (left panel, at  $t = 277.22$  sec) leads to a renewal of logarithmic decay in stress (right panel). In the framework of DRT, this occurs as a renewal of the fastest relaxing elements. (b) Simulated two-step relaxation where the driving protocol is held constant and diffusion between elements is varied. The fitting function is the same as in Fig. 4(a).

series of logarithms as in Fig. 4(a), and the fitting parameter  $C$  grows from 1 in the absence of diffusion to as large as 3 for the specific  $(t_w^{(1)}, \Delta\epsilon^{(1)})$  two-step protocol simulated. With faster stress diffusion, the value of  $C$  decreases until eventually the nonmonotonicity vanishes.

The diffusion of stress between relaxers lies in the same vein as modifications to DRT where the relaxation rates can evolve in time [27]. It offers a means to incorporate coupling between elements and to venture into the gulf between parallel and sequential models of relaxation (e.g., Refs. [28–30]). However, we also find that this implementation of diffusion modifies single step relaxation away from logarithmic at large timescales, and preliminary sweeps of three-step relaxation did not lead to values of  $C$  much greater than 2.

In this work, the seemingly paradoxical behavior of a nonmonotonically relaxing disordered system has been investigated in a new guise: a granular packing of glass spheres. The general DRT model, with the AOI distribution of relaxation rates, was used to explain two-step relaxation and predict three-step relaxation before exposing its own deficiency in the fitting function for the stress relaxation. Three-step relaxation experiments thus present the most stringent test to date for models of anomalous relaxation, and offer unique insight to the dynamics of far-from-equilibrium systems.

We thank Tom Witten, Sidney Nagel, Shmuel Rubinstein, and Thomas Videbæk for insightful discussions. This work was supported by the National Science Foundation through Grant No. CBET-1605075 and by the Chicago Materials Research Science and Engineering Center through Grant No. DMR-1420709. K. M. acknowledges support from the Center for Hierarchical Materials Design (CHiMaD).

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