Laser Speckle Strain Imaging reveals the origin of delayed fracture in a soft solid

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Stresses well below the critical fracture stress can lead to highly unpredictable delayed fracture after a long period of seemingly quiescent stability. Delayed fracture is a major threat to the lifetime of materials, and its unpredictability makes it difficult to prevent. This is exacerbated by the lack of consensus on the microscopic mechanisms at its origin because unambiguous experimental proof of these mechanisms remains absent. We present an experimental approach to measure, with high spatial and temporal resolution, the local deformations that precipitate crack nucleation. We apply this method to study delayed fracture in an elastomer and find that a delocalized zone of very small strains emerges as a consequence of strongly localized damage processes. This prefracture deformation zone grows exponentially in space and time, ultimately culminating in the nucleation of a crack and failure of the material as a whole. Our results paint a microscopic picture of the elusive origins of delayed fracture and open the way to detect damage well before it manifests macroscopically.

INTRODUCTION

Loading a solid with a stress below its critical fracture stress does not mean that the solid is safe from catastrophic failure. Subcritical stresses can lead to the sudden and unpredictable fracture of the material after a certain time delay, during which no precursors of the imminent failure are discernible by conventional imaging methods. Delayed fracture is observed in a wide variety of solids, ranging from brittle solids, such as ceramics (1), to ductile metals (2), heterogeneous (porous) media (3, 4), plastics (5), two-dimensional crystals (6), and a range of soft materials (7–11). Depending on the ratio of the applied load to the critical fracture stress, where crack nucleation is instantaneous, the delay time between loading and failure can vary from seconds to hours, days, or even years (1, 9, 10, 12). The combination of the wide range of possible delay times and the absence of visible precursors to the failure makes delayed fracture difficult to predict. Consequently, this failure mode poses a significant threat to the operational safety and lifetime of a wide variety of common materials. Because the microscopic mechanisms governing the time delay between stress application and the moment of catastrophic failure remain elusive, its prevention has remained highly challenging.

Contrasting explanations have been put forth to explain delayed fracture. Explanations can be divided into two main categories. First, delayed crack nucleation has been hypothesized to be caused by the thermally activated emergence of a flaw larger than the critical Griffith length (3, 4, 7, 13). By contrast, the erosion hypothesis proposes that the time delay nucleation of a crack as the result of a self-accelerating cascade of molecular damage processes at a site of stress localization. These damage processes erode the rigidity of the solid to zero upon which a crack can nucleate without an energy barrier (8–10, 14). In the first scenario, the scaling of the time delay \( t_d \) between loading and failure is predicted to scale with stress \( \sigma \) as \( t_d \propto \exp(-\sigma^a) \) (13), whereas the second theory predicts \( t_d \propto \exp(-\alpha) \) (8, 14). Because of the inherent time limitations of macroscopic mechanical experiments, distinguishing accurately between these two steep dependencies is virtually impossible. Rather, the definitive answer to the origins of delayed failure may be found microscopically: Whereas the first scenario predicts no precursors to fracture, a cascade of damage processes and resultant damage-induced deformations to maintain a local force balance must precede crack nucleation in the second. To date, the lack of experimental approaches to assess these nanoscopic strains deep within a material at a locus of stress concentration in the moments before fracture has precluded the resolution of this debate.

Here, we use Laser Speckle Strain Imaging (LSSI) to create highly resolved micromechanical maps of an elastomer in the moments before the nucleation of a delayed crack. In this way, we quantitatively probe the small but distinct emergence of a zone of diverging strains that are the result of strongly localized bond rupture events. The deformation field is centered at the locus where a crack will nucleate once the damage is complete; the emergent strain zone is found to grow exponentially and reaches macroscopic dimensions until failure is inevitable. Our experiments provide strong evidence—supported by spring network simulations—for the erosion mechanism of delayed failure, in which crack nucleation is preceded by small-scale damage that results in large-scale strains in the prefracture zone.

RESULTS

Visualizing the strain precursors to crack nucleation

To resolve the local strains in a rubber en route to delayed fracture, we create quantitative micromechanical maps using LSSI (for details, see Materials and Methods) (15–22). We study a prenotched, silica-filled silicone elastomer, which exhibits pronounced delayed failure after a period of apparent stability (Fig. 1A). Some weak visco- and poroelastic relaxation is observed just after application of a step strain because of solvent migration and viscoelastic deformation (11, 23). Because there is a clear separation of time scales between this relaxation and the delayed fracture, we treat these as independent phenomena. The nucleation of a delayed crack occurs with no discernible precursors in the macroscopic mechanical signal. To probe whether precursors to the failure can be detected microscopically, we add 0.2 weight % (wt %) of strongly scattering titania nanoparticles to the elastomer as tracers. We quantify the internal dynamics in the elastomer during the mechanical experiment in space and time by analyzing the fluctuations of the speckle pattern created by multiply scattered photons (Fig. 1, B and C, and...
concentrated near the notch, causing the scission of covalent bonds to initiate there; this bond breakage is highly anisotropic in filled rubbers (31, 32). Because the macroscopic strain is fixed, a change in the local elasticity due to bond rupture must require a redistribution of local stresses, hence local strains, to accommodate for the mechanical erosion that precedes nucleation. The emergent zone of enhanced and accelerating strains is thus a large-scale manifestation of the damage precursors, predicted by the erosion hypothesis (8, 9, 14), that occur much more locally.

**Validation of experimental methodology**

Before we quantify the mechanical precursors to crack nucleation in more detail, we aim to establish the validity and accuracy of our interpretation of the measurement data. To do so, we perform several reference experiments. First, we explore different contributions to the detected dynamics. Plots of contrast function \(d_2\) as a function of distance from the notch \(\Delta x\) demonstrate that, at the correlation time that we use, \(\tau = 0.5\) ms, the possible sources of speckle decorrelation are well separated (Fig. 2A): The noise threshold of our LSSI setup is negligibly small (♦), whereas the intrinsic thermal fluctuations of nanoparticles in a quiescent and stable elastomer (■ and ▲) produce a significantly smaller decorrelation than the dynamics resulting from damage-induced deformations (♦). Thus, the signal from the emergent strains that we probe is an order of magnitude larger than the background thermal fluctuations of the tracer particles and two orders of magnitude above the noise level.

Two types of particle motion contribute to signal decorrelation in these LSSI experiments. First, the embedded nanoparticles exhibit Brownian motion within the elastomer, characterized by diffusive statistics with a mean-square displacement \((\Delta x^2)\) that is limited because of the “cages” imposed by the cross-links (33). Second, localized damage processes within the solid alter its elasticity and result in stress redistribution and resulting strains. In principle, the \(d_2\) function measures a convolution of both types of motion. To disentangle these contributions, we first convert \(d_2\) to the electric field correlation

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**Fig. 1. Imaging local prefracture deformations.** (A) Macросopic stress transient in a step-strained notched elastomer en route to delayed fracture (denoted by “x”). (B) Schematic illustration of forward-scatter LSSI used to visualize strain precursors within the solid. The elastomer is illuminated with a coherent plane light wave, which is multiply scattered from TiO2 nanoparticles embedded in the elastomer, giving rise to (C) speckle patterns on a camera on the opposite side. Spatiotemporal correlation of the speckle intensity fluctuations, using the \(d_2\) function (Eq. 1), is used to create image contrast, encoding the local dynamics. (D) The resulting mobility maps reveal a growing zone of nanoscopic deformations, culminating in (E) catastrophic failure and the nucleation of a brittle crack. The background is masked in all images.
earlier work (sensible correlation time strain between two correlated speckles (with high resolution. In the remainder of the article, we will refer to evaluate changes in the amplitude of the local deformation field cannot provide an absolute measure for the strain, it provides a means

limit of multiple scattering, with a ratio of sample thickness as
solution of displacements that we can detect with reasonable accuracy as
the data, validating our analysis procedure. Inset: Average of different components of the strain tensor; therefore, we use \( \sqrt{f(U)} \) as a measure for the differential strain between two correlated speckles (34), here for the shortest possible correlation time \( \tau = 0.5 \text{ ms} \). Although this one-point correlation cannot provide an absolute measure for the strain, it provides a means to evaluate changes in the amplitude of the local deformation field with high resolution. In the remainder of the article, we will refer to \( \sqrt{f(U)} \) as the “differential strain intensity.”

The validity of this approach has been extensively demonstrated in earlier work (15, 20, 34–36), yet only in the backscatter geometry that is biased toward surface strains. Our forward-scatter setup offers two advantages: It probes the strains in the bulk of the material and offers a larger sensitivity to small deformations. As this method operates in the limit of multiple scattering, with a ratio of sample thickness \( h \) to \( l^0 \) of \( \sim 100 \), angstrom-scale motion of the scatterers already elicits a significant decorrelation of the speckle intensity. We estimate a minimum resolution of displacements that we can detect with reasonable accuracy as small as \( \delta_{\text{disp}} \approx 0.2 \text{ nm} \). This excellent sensitivity gives us the ability to measure the extremely small differential strains preceding delayed fracture.

To further verify our method as a means to quantify local strains, we subject an unnotched elastomer to a constant tensile strain rate \( \dot{\epsilon} = \text{d}e/\text{d}t \) and compute the differential strain intensity in the center using Eq. 2. Divided by the time separating the two correlated speckle patterns, here \( \tau = 5 \text{ ms} \), \( \sqrt{f(U)}/\tau \) is analogous to the local strain rate. We find a linear relationship between the measured local strain rate and that applied macroscopically (Fig. 2B). The two are related by a geometry-dependent proportionality constant, which results from the fact that \( f(U) \) is a scalar invariant of the strain tensor through a combination of different components of the tensor. The validity of our analysis is further supported by the identical shapes of the \( d_2(\tau) \) curves for different strain rates (Fig. 2B, inset), which are only shifted on the time axis. Finally, we note that the accuracy of the measurement is good: The SDs of time averaging \( d_2 \) over a constant \( \dot{\epsilon} \) period of 2 s are very small (Fig. 2B, inset), and those of spatially averaging \( d_2 \) over an area of 4.5 \text{ mm}^2 are also less than 10% in all cases.

For these reference samples subjected to a constant strain rate in the linear regime, the local deformation field is highly uniform. However, before a delayed fracture, we observe how a strongly heterogeneous deformation zone emerges (Fig. 1D). To establish the accuracy of the \( d_2 \) images in Fig. 1D, which we interpret as micromechanical maps of the elastomer during its mechanical perturbation, we must take into account the effects of spatial blurring. The measured signal in LSSI measurements is inherently a convolution of the local dynamics and the spatial distribution of photons in the material. Using random-walk simulations (37, 38), we find that the lateral blurring has a decay length of the order of the sample thickness (fig. S2). Thus, in these experiments, we inevitably deal with a trade-off between spatial resolution and strain detection sensitivity: Although the ratio of \( h/l^0 = 100 \) provides high strain sensitivity; it also results in spatial blurring due to the long photon paths. Despite this spatial blurring, the observed changes in \( f(U) \) with time are significant.

Quantification of local deformation field
Having established the validity and limits of our method, we now proceed to explore the prefracture deformation zone by plotting \( \sqrt{f(U)} \) as a function of the time to fracture \( \Delta t \) in the last 7.7 s before fracture, for different distances \( \Delta x \) from the notch. These data show the emergence

\[
g_1 \approx \frac{k_0 h \sqrt{3f(U) + \langle \Delta r^2 \rangle/l^2}}{\sinh \left[ k_0 h \sqrt{3f(U) + \langle \Delta r^2 \rangle/l^2} \right]} \tag{2}
\]

where \( k_0 = 2\pi n/\lambda \) is the wave vector with \( n = 1.5 \) the refractive index of poly(dimethylsiloxane) (PDMS), \( h = 5 \text{ mm} \) is the sample thickness, \( f(U) \approx \frac{1}{\bar{U}} \left[ Tr^2(U) + 2Tr(U^2) \right] \) with \( U \) the strain tensor and \( Tr \) its trace, \( l^0 = 50 \mu \text{ m} \) is the photon transport mean free path determined independently by means of coherent backscattering (see Materials and Methods), and \( \langle \Delta r^2 \rangle \) is the Brownian mean-square displacement of the tracer particles. We consider the latter constant (at fixed \( \tau \)) and set it at the mean-square displacement of the nanoparticles in the quiescent material (see Materials and Methods). Because elastomers are nearly incompressible, with Poisson’s ratio \( \nu \approx 0.5, \) \( Tr(U) \) is negligible (15), yielding \( f(U) \approx \frac{1}{\bar{U}} Tr(U^2) \). The complexity of our experimental geometry precludes computation of the individual components of the strain tensor; therefore, we use \( \sqrt{f(U)} \) as a measure for the differential strain between two correlated speckles (34), here for the shortest possible correlation time \( \tau = 0.5 \text{ ms} \). Although this one-point correlation cannot provide an absolute measure for the strain, it provides a means to evaluate changes in the amplitude of the local deformation field with high resolution. In the remainder of the article, we will refer to \( \sqrt{f(U)} \) as the “differential strain intensity.”

Fig. 2. Setup sensitivity and strain quantification. (A) Comparison of the average nanoparticle mobility \( d_2 \) in a thin horizontal strip in four different samples: an elastomer strained by 25% at \( \Delta x = 1 \text{ s} \) (●), an elastomer strained by 3% (▲), the same elastomer in its quiescent, unstrained state (■), and a static opaque medium, that is, a piece of ground glass, in which no thermal speckle fluctuations occur (▲). The notch is located at \( \Delta x = 0 \). The angular brackets denote averaging over 40 pixels in the orthogonal direction. (B) Apparent local strain rate in the center of an elastomer strained at a constant macroscopic rate. The dashed line is a predicted linear fit to the data, validating our analysis procedure. Inset: Average \( d_2(\tau) \) curves from which \( \sqrt{f(U)} \) is computed, here at \( \tau = 5 \text{ ms} \) marked by the dotted line. The error bars indicate the SD of time averaging \( d_2 \) over 2 s.
of a growing deformation zone (Fig. 3A) that starts at the notch, where the initial stress concentration reaches a maximum. The local differential strain grows more steeply as the moment of failure approaches, at which it converges to a single value for all locations in the field of view. The exponential growth of $\sqrt{f(U)}$, which is the consequence of the molecular damage at the notch that underlies the delayed crack nucleation, is evidence for the erosion hypothesis because it indicates a self-catalytic process that accelerates over time.

We observe the same kinetics for a (higher) strain of 30% but on time scales that are two orders of magnitude shorter (Fig. 3B), which is in line with the scaling of the delay time with the applied macroscopic stress or strain. This further confirms our theory that the prefracture deformation zone is a slave of local damage processes: Such a steep dependence on the macroscopic strain is in accord with the picture of bond rupture as a Kramers process, in which bond breaking is thermally activated and exponentially enhanced by a mechanical load (8, 12, 39).

For both strains, our results illustrate how the precursors to crack nucleation expand in space at a constant rate but diverge steeply just before fracture. Specifically, by plotting the time at which the strain nucleation expands in space at a constant rate but diverge steeply just before fracture. By plotting the time at which the strain starts to rise before fracture. Specifically, by plotting the time at which the strain starts to rise $\Delta t$, for different distances from the notch, we find a linear increase in the first growth stage (Fig. 3C), whose slope is a measure for the spatial growth velocity of the deformation zone. Note that these velocities—0.17 and 5.6 mm/s for the 25 and 30% strained elastomer, respectively—are three orders of magnitude lower than the crack propagation rates—0.7 and 10 m/s—which are again two to three orders of magnitude lower than the speed of sound in the material (~km/s) (40). Clearly, the damage processes that precede failure are governed by different physics than those that dictate the propagation rate of a nucleated crack.

The kinetic processes underlying the prenucleation damage and resultant deformations show a remarkably similar evolution at all distances $\Delta x$ from the notch. We are able to collapse all 550 measured differential strain $\sqrt{f(U)}$ profiles onto a single master curve through rescaling the time axis with a strain- and distance-dependent factor $k(\varepsilon, \Delta x)$, which represents the rate with which the local strain accelerates (Fig. 3D). The absolute values of $k$ for the 25 and 30% strained elastomers differ by two orders of magnitude (Fig. 3D, inset), in line with the picture of force-enhanced bond rupture and rigidity erosion. However, their dependence on $\Delta x$ and $k$ first increases and then reaches a plateau value far from the notch. This form is very robust and shows a negligible dependence on the extent of (orthogonal) spatial averaging up to ~40 pixels per bin, with low fit errors in all cases (fig. S3). The initial increase arises from the expansion of the deformation zone and acceleration of the damage processes at its origin, causing locations farther away from the notch to experience a faster rise of $\sqrt{f(U)}$ toward the moment of crack nucleation $\Delta t = 0$. Exactly at that time point, all the differential strains in the field of view converge, marking the transition from small-scale

![Fig. 3. Localized growth of nanodeformations. (A and B) The local differential strain intensity $\sqrt{f(U)}$ increases exponentially toward the moment of crack nucleation $\Delta t = 0$, shown for an elastomer strained macroscopically by $\varepsilon = 25\%$ (A) and $30\%$ (B) as a function of the distance to the locus of highest stress concentration: $\Delta x = 0.1, 0.28, 0.4, 0.55$, and 1.5 mm. The angular brackets denote averaging over 40 pixels in the orthogonal direction [see inset in (A)]. The expansion of the precursor zone is evidenced by the different starting points $\Delta t_0$ of the $\sqrt{f(U)}$ curves. (C) Spatial dependence of $\Delta t_0$ for a macroscopic strain of $\varepsilon = 25\%$ (right ordinate) and $30\%$ (left ordinate). The initial linear growth is marked by the fitted solid lines, whose slopes reveal growth velocities of 0.17 and 5.6 mm/s, respectively. (D) Collapse of all transient $\sqrt{f(U)}$ profiles, illustrating universality, through rescaling of the time axis with the local strain acceleration rate constant $k(\varepsilon, \Delta x)$ (see inset). Ten percent of all data points are shown.](image)
deformations to macroscopically detectable strains involved in the catastrophic failure.

**Simulating delayed fracture**

Finally, to strengthen the proposed connection between localized molecular damage and delocalized deformations, we perform simulations of a spring network model \((41, 42)\) subjected to a step strain. To describe damage accumulation, we irreversibly soften the bonds in the network when the force on them exceeds a critical rupture threshold \(f_{thr}\). Each bond is allowed to soften a maximum of \(n_{soft}\) times before it is broken. The order in which the bonds are softened and ultimately broken is deterministic with the weakest bond damaging first, yet two stochastic rules are implemented in the simulations: (i) Local rearrangements are implicitly taken into account by randomly generating a new \(f_{thr}\) after each event \((42, 43)\), according to the current stiffness \(m_{bin}\). Softer regions have a higher probability of becoming weaker. (ii) The simulation time \(t^*\) is advanced in a stochastic fashion while additionally assuming a total rate constant \(R_{tot} \sim 1/dt^*\) that is proportional to the sum of the excess force (above \(f_{thr}\)) carried by all bonds \((44)\). Therefore, \(R_{tot}\) is a measure for the rate of damage accumulation, with a large \(R_{tot}\) implying fast damage dynamics, thus providing insight into the molecular bond scission processes.

In our simulations, we observe the localization, accumulation, and diffusion of damage preceding crack nucleation (Fig. 4). Notably, the triangular shape of the damage zone (Fig. 4, A to C) resembles the shape of the deformation zone observed in experiments and is independent of the choice of lattice topology and simulation parameters, within reasonable bounds (see fig. S4 and the Supplementary Materials for details). Despite the differences between simulations and experiments in the length scales, the number of bonds involved in the failure process,
and in the network heterogeneity, our simulations provide further evidence that the deformations measured experimentally can be directly related to bond rupture events close to the notch. Bonds break as a direct result of the stress concentration imposed by the notch geometry; the consequent stress redistribution and growth of the damage zone cause deformations of springs progressively farther from the notch (Fig. 4, E to H). Future effort should be dedicated to improving the microscopic model to allow a quantitative comparison with experiments; for example, the delay time and the size of the damage zone can be tuned by subtly varying the softening-breaking protocol (figs. S5 and S6).

**DISCUSSION**

Our results shed new light on a long-standing debate regarding the mechanisms of delayed failure, ruling out the hypothesis of subcritical crack nucleation as a sudden event without any precursors. Instead, we find that, well before the fracture is macroscopically detectable, a distinct zone of small-scale deformations emerges at the exact locus of the ultimate crack nucleation. This local strain field grows exponentially in time and space and follows similar kinetics as the bond rupture cascade predicted by the erosion hypothesis. Because the resolution of LSSI is too low to directly visualize bond rupture, other techniques are required to provide conclusive evidence. Nevertheless, all our findings are in line with delayed fracture being the result of strongly localized damage accumulation, which concomitantly causes delocalized deformations.

The originality of our methodology lies predominantly in the high deformation resolution, which far exceeds that of commonly used techniques to measure surface strains such as digital image correlation. In addition, LSSI not only provides information about the static deformation field but also offers a wealth of dynamic information that allows different processes to be disentangled and quantified separately. The full capacity of this technique is still far from fully explored. For instance, the results in this paper suggest that LSSI may pave the way to early detection of damage in existing materials well before its catastrophic effects become manifest. Moreover, it can provide a deeper understanding of the mechanisms of damage and dissipation in a much larger class of mechanical problems. Many challenging questions remain unanswered in the field of fracture and damage mechanics, in particular in those scenarios where the powerful continuum approach of linear elastic fracture mechanics breaks down (45), for example, experimentally probing the effects of heterogeneity that are predicted to fundamentally alter the physics of mechanical instabilities (46). Finally, with the emerging trend of ultrathin solids through the molecular design of dissipative pathways (30, 47, 48) or self-healing capabilities (49, 50), the need for techniques to elucidate the molecular dynamics underlying these unique mechanical properties has grown, and optical micromechanical mapping is a powerful tool to contribute to the rational design of tough and self-healing solids (21).

**MATERIALS AND METHODS**

**Materials**

We studied delayed fracture in single-edge-notched, cross-linked PDMS elastomers in which we embedded 0.2 wt % of TiO2 nanoparticles as scattering tracers. The elastomers were prepared by mixing PDMS Sylgard 184 (Dow Corning) prepolymer and cross-linking agent at a 5:1 weight ratio with 0.2 wt % TiO2 nanoparticles (coated with silicone oil; diameter, ~30 nm; US Research Nanomaterials); the low TiO2 nanoparticle concentration of 0.2 wt % ensured that particle–particle interactions are absent, while their coating with silicone oil precluded interactions with the PDMS matrix. Air bubbles were removed by centrifugation at 400g for 2 min. Subsequently, the mixture was poured into rectangular molds coated with a release agent (Universal Mold Release, Smooth-On). The molds were 60 × 15 × 5.0 mm3 (l × w × h) and featured a rectangular notch of 0.20 × 10 × 5.0 mm3 centered on the long edge. The samples were cured overnight at 65°C, resulting in highly opaque elastomers that are macroscopically brittle, with fracture toughness \( K_{IC} = 0.2 \) MPa \( \sqrt{m} \) measured by dynamic mechanical analysis (see subsection below) and Young’s modulus \( E = 1.1 \pm 0.1 \) MPa determined from the initial linear slope of a stress-strain curve (implying a mesh size on the order of \([K_{IC}/E]^{1/3}\) = nanometers). The photon transport mean free path was measured by coherent backscattering (51) to be \( \ell = 50 \) μm (see subsection below). It is important to note that Sylgard 184 contains at least 16 volume % of silica filler, in the form of ~100-nm aggregated silica particles (32), added by the manufacturer to increase the tear strength. The mechanical properties and structure of silica-filled PDMS networks have been studied extensively (32, 52).

**Forward-scatter LSSI**

We created quantitative micromechanical maps with high spatiotemporal resolution using a custom-built LSSI setup in the forward-scatter geometry. LSSI operates on the same optical principles as diffusing wave spectroscopy (53). In our experiments, the sample was placed in a strain-controlled tensile tester and illuminated with an expanded coherent light beam (1.5 W, \( \lambda = 532 \) nm; Cobolt Samba), orthogonal to which the elastomer was strained (Fig. 1B and fig. S1). The photons in the illumination beam were injected into the material and multiply scattered by the embedded nanoparticles. Because the sample thickness \( h = 5.0 \) mm was 100× larger than the transport mean free path \( \ell = 50 \) μm, the photons that exited the material in the forward-scattering direction had undergone numerous scattering events; consequently, they had traversed a diffusive rather than a ballistic trajectory within the sample. Each photon performed a unique random walk, giving rise to path length differences among the photons that caused random constructive and destructive interference in the scattered signal, which was recorded on a complementary metal-oxide semiconductor chip of a high-speed camera (with an acquisition rate of 2000 Hz; HiSpec 1, Fastec Imaging) as a speckle pattern of bright and dark spots. Both laser and camera were directed at the tip of the notch, where the tensile stresses were concentrated, dictating the site of eventual crack nucleation. Only the multiply scattered light was detected, with transmitted and low-order scattering paths filtered by a linear polarizer perpendicular to the polarization of the incident laser beam.

As the embedded nanoparticles underwent thermal fluctuations, all photon path lengths changed, causing the speckle pattern to change accordingly. The rate of these fluctuations was analyzed on a pixel-by-pixel basis using the intensity structure function \( d_2 \) (Eq. 1), which provides access to quantitative information about the internal dynamics within the material. The angular brackets in Eq. 1 denote averaging in time and/or space. Specifically, to create the \( d_2 \) images in Fig. 1D, the speckle fluctuations were averaged over 100 frames = 50 ms in time and an additional 3 × 3 pixels = 7.8 × 102 μm2 in space that acted as a sliding window. To compute the spatial and temporal \( \langle d_2 (\tau) \rangle \) and \( \sqrt{\langle f(U) \rangle} \) profiles in Figs. 2A and 3, the speckle fluctuations were averaged over 5 ms in time and an additional 40 pixels = 3.5 × 102 μm2 in space. Finally, the \( \langle d_2 (\tau) \rangle \) curves in Fig. 2B were obtained by multispeckle
averaging over a rectangular area of 300 × 170 pixels = 4.5 mm², followed by time-averaging the resulting d2 values over the first 2 s after application of the strain rate.

To allow computation of \[ \sqrt{f(U)} \] using Eq. 2, the contrast function \( d_2 \) was first converted to the electric field correlation function \( g_1 \) by \( g_1(\tau) = \sqrt{1 - d_2(\tau)/2\beta} \). Here, the Siegert relation, 
\[ g_1(\Delta t, \tau) = \sqrt{g_2(\Delta t, \tau) - 1}/\beta, \]

is applied and the fact that \( g_2(\Delta t, 0) = g_2(\Delta t, \tau) = 2 \) (54). The \( g_2 \) function itself can be directly computed from the raw speckle patterns, yet in this work, \( d_2 \) was consistently used as the starting point because \( g_2 \) is more reliable than \( g_1 \) in cases of relatively poor statistics (54). \( \beta \) is the spatial coherence factor that accounts for the number of speckles detected. It was chosen such that \( d_2(\tau) \rightarrow 2\beta \) for \( \tau \rightarrow \infty \).

The mean-square displacement in Eq. 2 was calculated by \( \langle \Delta r^2 \rangle = -6 \ln g_1/[k_0 h/f]^2 \). The local strain acceleration rate constant \( k(\Delta t, \Delta x) \) was obtained by least-squares fitting the differential strain intensity to an empirical, single-exponential equation
\[ \sqrt{f(U)}(\Delta t) = 1.7 \times 10^{-7} + 5 \times 10^{-6} \cdot e^{\Delta t}. \]

**Dynamic mechanical analysis**

The macroscopic mechanical properties of the elastomers were measured using a dynamic mechanical analyzer (Q800, TA Instruments). The samples were gripped using a film tension clamp with a clamp compliance of 0.24 mm/N, and uniaxial tensile tests were conducted at a controlled strain ramp rate of 20%/min. The Young’s modulus \( E \) was obtained from the initial slope of the stress–strain curves. The fracture toughness was computed as \( K_{IC} = \sigma_Y \sqrt{a} \) with \( \sigma_Y \) the critical fracture stress, \( a = 10 \) mm the notch length, and \( Y = [1.12 + a(2.91a/w - 0.64)/w]/[1 - 0.93a/w] = 5.2 \) the geometric configuration factor.

**Coherent backscattering**

The transport mean free path \( \lambda = 50 \) μm in the elastomers was estimated by measuring the decay of a coherent backscattering cone. See the study of Verma et al. (51) for a detailed description of the method. Briefly, the elastomer was illuminated with a 520-nm laser beam (Thorlabs MCLS1 4-Channel Laser Source), and a total of 500 images of the coherent backscattering cone were acquired with an electron-multiplying charge-coupled device camera (Hamamatsu C9100-13) to ensure ample statistics. The image intensities were azimuthally averaged to obtain the radial intensity decay curve, which was fitted to extract \( \lambda \) as described in the study of Verma et al. (51).

**Spring network simulations**

A network of linear elastic springs \( f_i = \mu_i [l_i - l_0] = \mu_i \Delta l_i \) with identical stiffness \( \mu \) and rupture force threshold \( f_{thr,i} \) was initially prepared by placing \( N_x \times N_y \) nodes on a triangular lattice with spacing \( l_0 \) and featuring vertical bonds. A notch was placed in the network by removing the nodes inside a rectangular region (width \( w_n \) and height \( 2r_{notch} \)) with a circular end of radius \( r_{notch} \). A step strain was applied to the network, and all the nodes were displaced affinely. Nodes on the top, bottom, and right boundaries were kept fixed. The network was relaxed by minimizing the total elastic energy, and the internal nodes were free to move off-lattice; the softening-breaking protocol was subsequently applied. The energy minimization was performed using the fast inertial relaxation engine (FIRE) algorithm (55) with tolerance \( F_{rms} = 10^{-5} \) (in reduced units) and the other parameters as suggested in the original paper.

The weakest bond was identified as the one with the largest excess \( f_i f_{thr,i} \) and underwent softening \( \mu_i \to 0 \mu_i \). The energy was minimized again, and the procedure was repeated. Each bond was allowed to soften a maximum of \( n_{soft} \) times before it was broken. After each softening, a new rupture threshold for the bond was randomly generated. In particular, after the \( j \)th softening the threshold was uniformly picked in \( [f_{min,i j}, f_{thr,i}] \) with \( f_{min,i} = a f_{thr,i} \).

The simulation quantities were expressed in reduced units by setting the lattice spacing \( l_0 = 1 \) and the initial spring constant \( \mu = 1 \) and by assuming a linear proportionality between rate constant \( r_i \) and excess force \( f_i/f_{thr} > 1 \) that sets the units for the simulation time \( t^* \). The time evolution of the system was therefore dictated by the rate equation
\[ R_{tot} = \Sigma_i r_i = \Sigma_i f_i/f_{thr,i} \]

where the sum extends to all springs \( i \) with \( f_i/f_{thr,i} > 1 \). The time interval between two softening-breaking events was drawn randomly assuming exponentially distributed events:
\[ dt^* = -\ln (\text{rand}(0, 1))/R_{tot}. \]

We defined \( (\Delta x_{dam}^i) = [\sum_{l=1}^{N_{dam}} \Delta x_i d(l)/N_{dam} \] with \( N_{dam} \) as the number of damaged springs, \( d_i = 1, \ldots, n_{soft} \) as the number of times that spring \( i \) has undergone softening, and \( \Delta x_i \) as its distance from the notch. The stiffness in the regions close to the notch was obtained by summing the spring constants in bins of width \( 3l_0 \times 5l_0 \). The spring deformation rate was computed as \( [\Delta l_i(t + \Delta t)^2 - \Delta l_i(t)^2]/\Delta t \), with \( \Delta t \) the time between the snapshots that are 50 events apart. In Fig. 4 (E to H), springs are colored green when this rate exceeds the threshold \( 10^{-3} l_0^2/\mu^* \).

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/4/5/eaar1926/DC1

Supplementary discussion of simulations

fig. S1. Schematic top view of LSSI in the forward-scatter geometry.

fig. S2. Spatial blurring in forward-scatter LSSI.

fig. S3. Robustness of the fitting procedure of the \( \sqrt{f(U)}(\Delta t) \) profiles.

fig. S4. Simulation protocol and shape of damage zone.

fig. S5. Tunability of the softening-breaking protocol.

fig. S6. Simulations at different strains for various notch radii.

movie S1. Specule movie of the damage-induced deformations preceding delayed fracture of a 25% strained elastomer.

movie S2. \( d_j \) movie of the growth of the prefailure deformation zone in a 25% strained elastomer.

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