

Supplementary Information for “Singular dynamics in the failure of soft adhesive contacts”

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SILICONE GEL SUBSTRATE PREPARATION

As in our earlier work [1–3], we make soft, solid silicone gels by combining a silicone base: vinyl-terminated polydimethylsiloxane (DMS-V31, Gelest Inc) with a cross-linker: trimethylsiloxane terminated (25-35% methylhydrosiloxane)-dimethylsiloxane copolymer (HMS-301, Gelest Inc). The ratio of polymer to cross-linker determines the stiffness of the cured silicone gel. The reaction is catalyzed by a platinum-divinyltetramethyldisiloxane complex in xylene (SIP6831.2, Gelest Inc).

To make the silicone, we prepare two parts: Part A consists of the base with 0.05 wt% of the catalyst. Part B consists of the base with 10 wt% crosslinker. We mix parts A and B together in a ratio of 9:1 by weight. The parts are mixed together thoroughly and degassed in a vacuum. While the silicone is still liquid, we prepare the experimental substrates in the desired geometry as described in the main text, and then cure the silicone in an oven in air at 70°C for at least 24 hours.

We measure the stiffness of the resulting gels by preparing bulk (all dimensions ≥ 1.5 cm) samples and measuring force F vs. indentation depth d while indenting with 3-mm-diameter cylindrical flat punch tool on a TA.XT Plus Texture Analyzer (Texture Technologies). We relate these measurements to the material elastic constants using Hertz indentation theory, such that

$$F = 2aE^*d = \frac{2aE}{1-\nu^2}d, \quad (1)$$

where $a = 1.5$ mm is the contact radius, E is the Young modulus, and $\nu = 0.48$ is the Poisson ratio, measured previously using compressive tests on a rheometer [1].

POWER-LAW RHEOLOGY

As discussed in the main text, we measure the shear rheology of the A:B 9:1 silicone gel with a small-strain (1%) frequency sweep using a 50 mm parallel plate geometry on a rheometer (Anton Paar MCR 502). We plot the results in Figure S1.

RAW DATA MOVIES

We include all raw data analyzed for this work as movies in .avi format:

Supplementary movies 1a & 1b: Detachments from a 5.0 kPa gel filmed at 500,000 fps, replayed at 5 fps

Supplementary movie 2: Detachment from a 5.0 kPa gel filmed at 220,000 fps, replayed at 10 fps

Supplementary movie 3: Detachment from a 5.0 kPa gel filmed at 78,000 fps, replayed at 10 fps

Supplementary movie 4: Detachment from a 5.0 kPa gel filmed at 3200 fps, replayed at 5 fps

Supplementary movie 5: Detachment from a 5.0 kPa gel filmed at 300 fps, replayed at 5 fps

Supplementary movie 6: Detachment from an 8.5 kPa gel filmed at 220,000 fps, replayed at 10 fps

Supplementary movie 7: Detachment from an 8.5 kPa gel filmed at 78,000 fps, replayed at 10 fps

Supplementary movie 8: Detachment from a 17 kPa gel filmed at 220,000 fps, replayed at 10 fps

Supplementary movie 9: Detachment from a 17 kPa gel filmed at 78,000 fps, replayed at 10 fps

Supplementary movie 10: Detachment from uncrosslinked liquid PDMS (Gelest DMS-V31) filmed at 78,000 fps, replayed at 10 fps

BRIGHT-FIELD IMAGE PROFILE MAPPING

We map the profiles in the bright-field videos of the substrate iteratively by frame, using the intensity gradient of each image. We determine pixel to micrometer ratio with a stage micrometer. Figure S2 shows the steps

involved in the mapping process, starting with an original image in the first panel.

The mapping procedure we use is modified from our earlier work [1]. The modification is required because when stretched to a conical shape at early times, the transparent silicone can act as a lens for the illuminating light. The result of this lensing phenomenon is that the center of the profile appears bright, even though there is silicone material there. The edge itself remains dark, but the nearby lensing can sometimes cause errors in fitting for the precise edge position. To mitigate this effect, in the early frames of the videos where there was significant lensing, we first filled in the pixels well below the edge of the profile. An example of this filling is shown in Figure S2(2). We found that after about 400-500 microseconds, the profiles were flat enough that the lensing disappeared, so we no longer filled in below the edges after that time. We further compared mapped profiles with and without the fill, and found no difference in the measured positions other than when the lensing destabilized the profile fit.

We then slightly smooth the raw bright-field image to reduce noise in the edge finding, shown in Figure S2(3). For the higher frame rate experiments, we smooth using a 3-by-3 pixel averaging filter to ensure that we preserve the early-time peaks; for lower frame rate experiments, we smooth with a 6-by-6 pixel filter to better resolve small deformations.

After smoothing, we take the gradient of the image using the MATLAB built-in function `imgradient()`, as shown in Figure S2(4). Finally, we map the edge profile by walking the curve that represents the image gradient maximum as measured by fitting normal to the profile (Figure S2(5)), using the same software as developed in our previous work [1]. We plot all mapped profiles from a series of experiments with $E = 5.0$ kPa substrate in Figure S3.

We used this method to map all detachment movies, except for the movies acquired at 500,000 frames per second. In this case, the field of view was too small to map the profile edge accurately. Instead, to analyze the 500,000 fps videos, we estimated the center of the peak in the horizontal (x) direction. Then, we directly fit the gradient of the image vertically at that x -position with a Gaussian to find the y -value of the peak position.

MEASURING DISTANCE FROM DETACHMENT AND HEIGHT VS. TIME

We measure the profile height, h , as the vertical position of the peak of each profile, extracted from the mapped profile data by fitting each profile with a smoothing spline. We measure the distance from detachment, δ , as the straight-line distance from the point of final contact to the peak of each mapped profile. The final contact point is calculated as the point on the sphere at the

same horizontal position as the center of the final profile before detachment. We estimate the error in these measurements as approximately the same as the profile mapping error, about 100 nm.

In all experiments, we capture a final image of the substrate in contact, followed by a clearly detached and recoiling surface in the next image frame. (See raw data movies, included in the ESI.) Consequently, there is an uncertainty of ± 0.5 frames in measuring the actual time of detachment, defined as $t = 0$. We estimate the real time of detachment as halfway between these frames. In order to explore the effect of this assumption on our measured power-law scalings, we plot δ vs. $(t \pm 0.5/\text{framerate})$ separately for each experiment in Figure S4. Similarly, we plot h vs. $(t \pm 0.5/\text{framerate})$ separately for each experiment in Figure S5. These plots suggest that our estimate of $t = 0$ as halfway between the last-attached and first-attached frames is reasonable.

EXPERIMENTS VARYING GEL STIFFNESS

For comparison, we performed detachment experiments on substrates of three different stiffnesses: $E = 5.0 \pm 0.1$ kPa, which experiments are the focus of this work, $E = 8.5 \pm 0.1$ kPa, and $E = 17 \pm 1$ kPa. We plot δ vs. t and h vs. t for experiments imaged at 220,000 fps on substrates of each stiffness in Figure S6.

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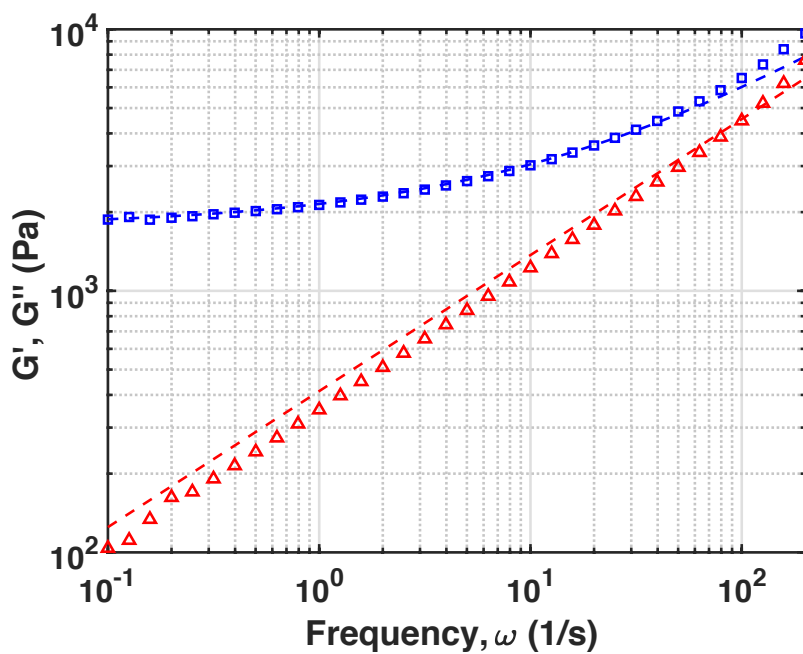


FIG. S1. Silicone gel rheology: Blue squares: G' , red triangles: G'' . Dashed lines show the fit to the more generalized framework of the Chasset-Thirion Equation [4, 5].

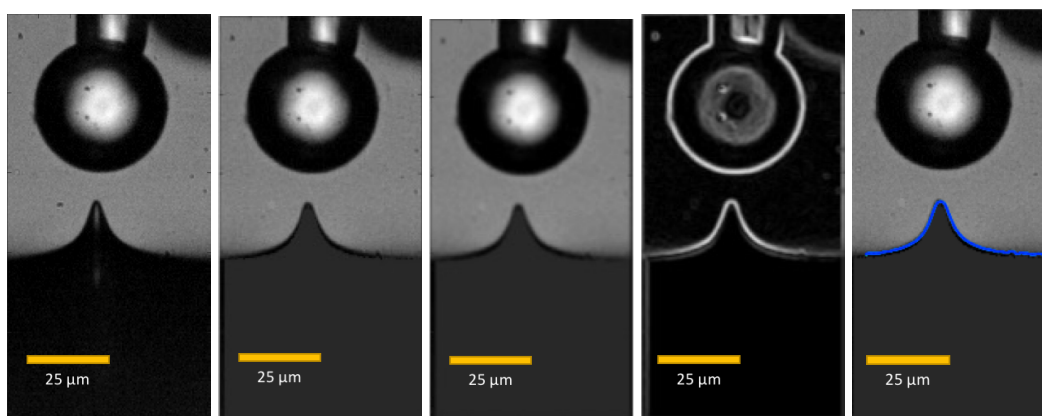


FIG. S2. Profile mapping procedure and result for an example substrate profile during post-detachment recoil. From left to right: (1) Raw bright-field image. (2) Original image, filled in dark well below the interface to remove extraneous bright spots from lensing. (3) Smoothed image. (4) Gradient of smoothed image. (5) Raw, filled image with mapped profile overlaid.

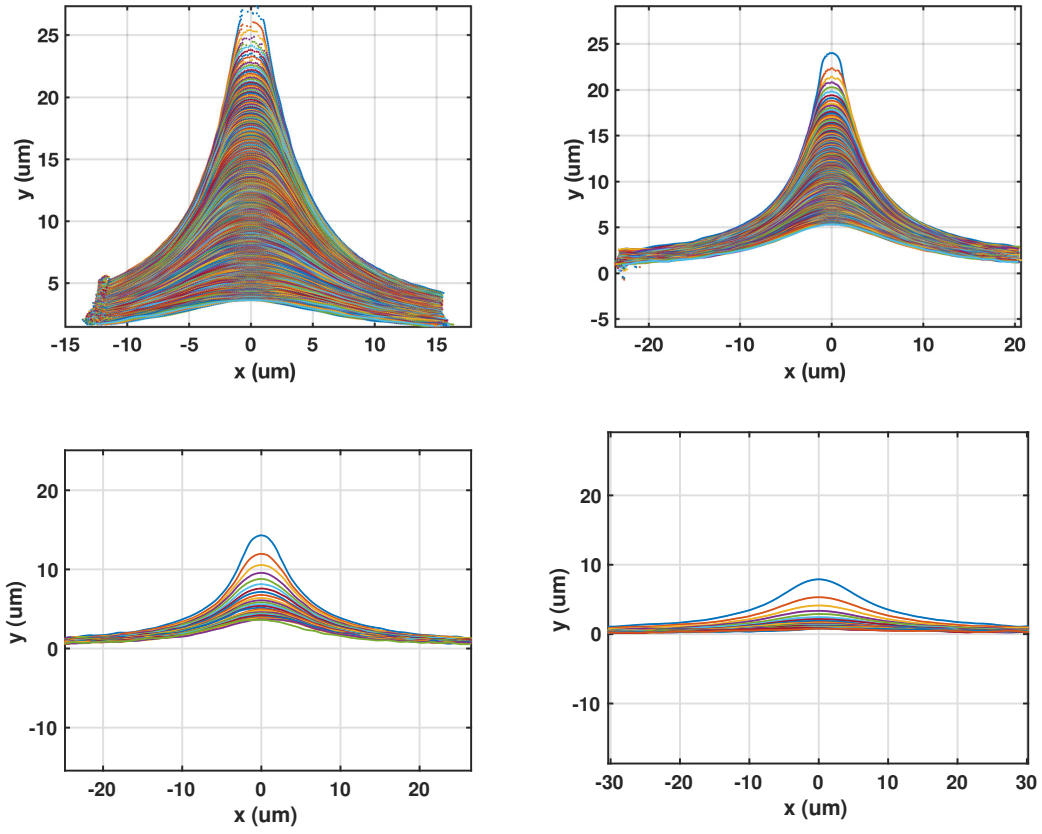


FIG. S3. Figures showing the all mapped profiles for experiments on a $E = 5.0$ kPa substrate, repeated at different frame rates. Upper-left: 220,000 fps; upper-right: 78,000 fps; lower-left: 3200 fps; lower-right: 300 fps. Profiles are zeroed vertically with respect to the undeformed surface, mapped from an image taken long after the post-detachment recoil had finished. The center of the axisymmetric profiles determines $x = 0$. These profiles form the basis for all further analyses.

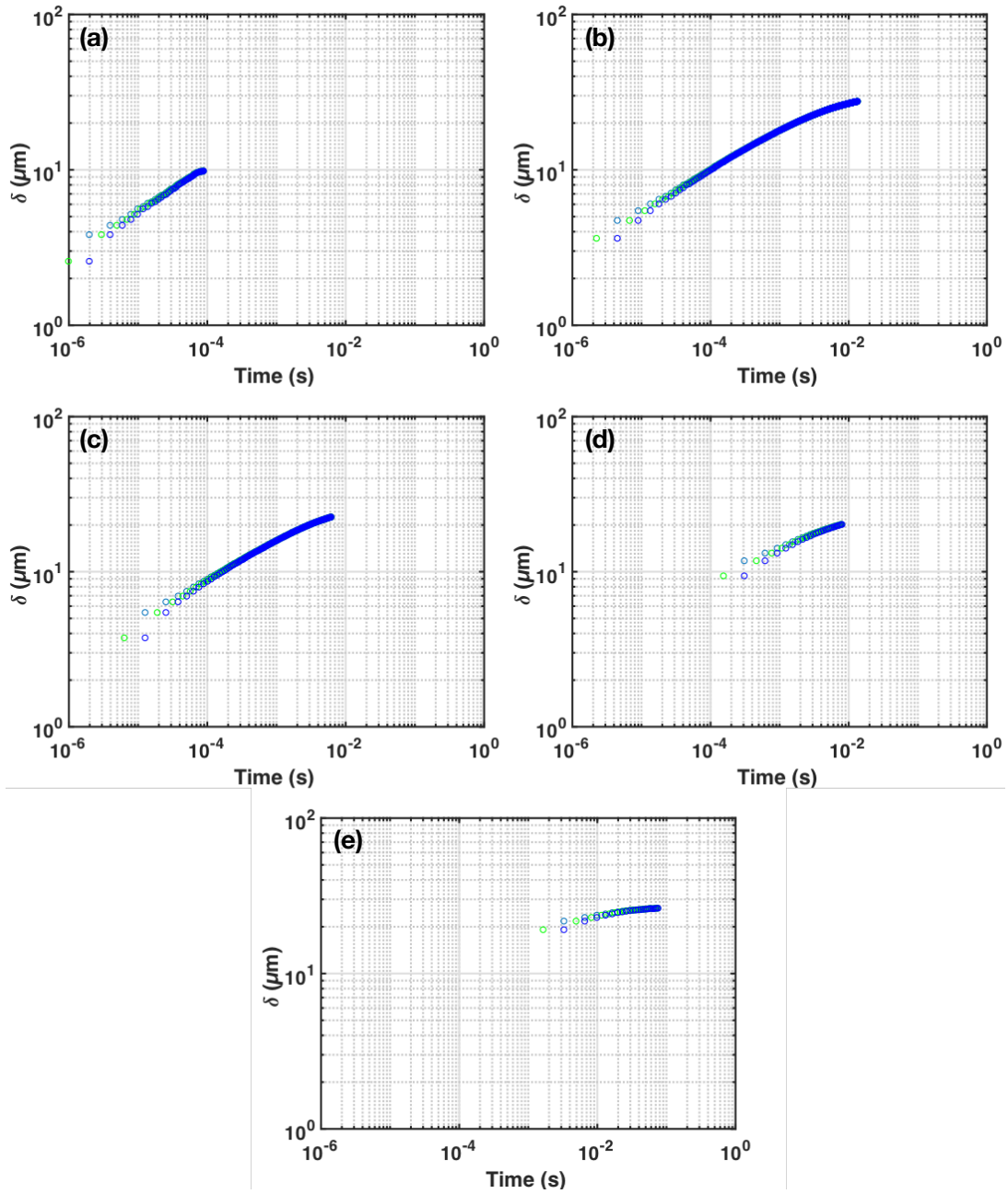


FIG. S4. Distance from detachment δ vs. time t for experiments imaged at: (a) 500,000 fps; (b) 220,000 fps; (c) 78,000 fps; (d) 3200 fps; (e) 300 fps. Each plot shows three results, corresponding to setting $t = 0$ to be (1) at the last attached frame, (2) at the first detached frame, and (3) halfway between the two.

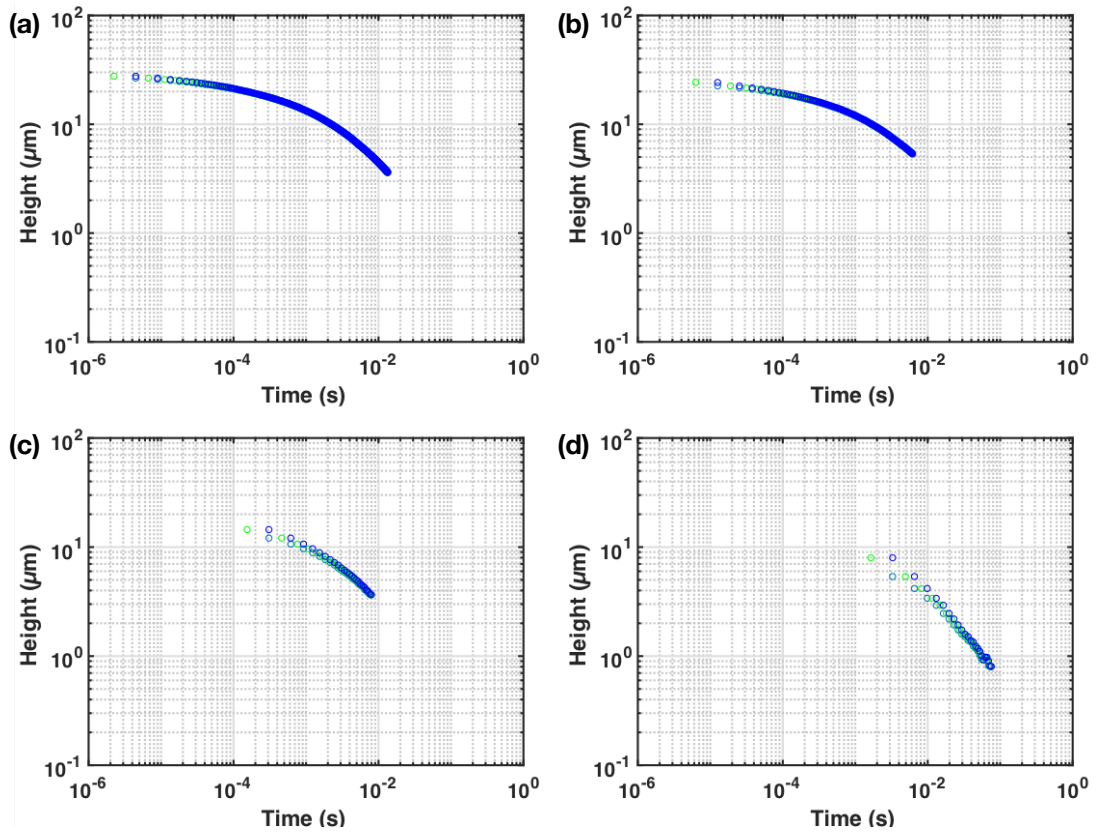


FIG. S5. Peak height h vs. time t for experiments imaged at: (a) 500,000 fps; (b) 220,000 fps; (c) 78,000 fps; (d) 3200 fps; (e) 300 fps. Each plot shows three results, corresponding to setting $t = 0$ to be (1) at the last attached frame, (2) at the first detached frame, and (3) halfway between the two.

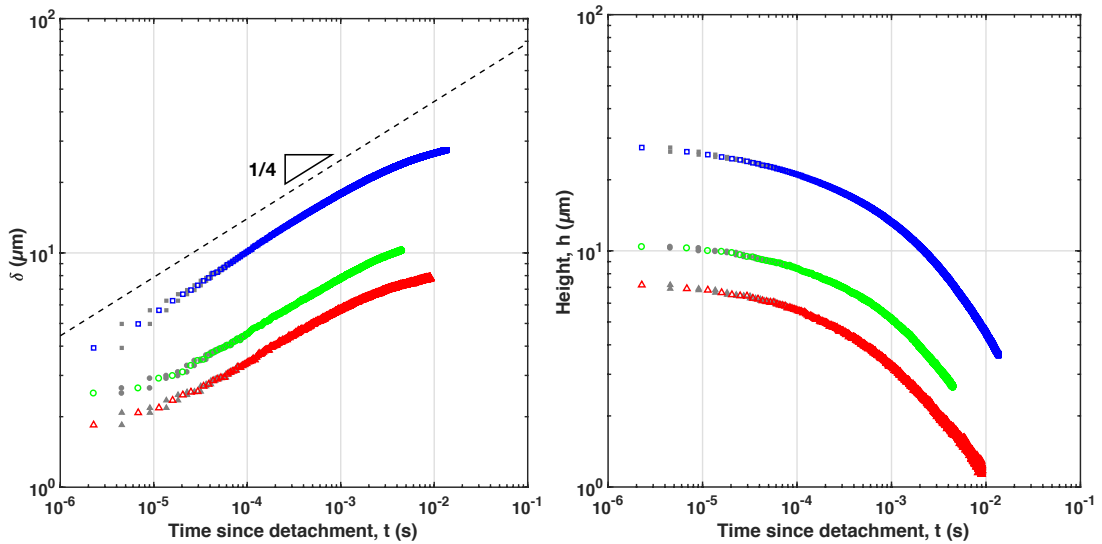


FIG. S6. Effects of varying stiffness. (left) Distance from detachment δ vs. time t for experiments imaged at 220,000 fps on substrates with $E = 5.0 \pm 0.1$ kPa (blue squares), $E = 8.5 \pm 0.1$ kPa (green circles), and $E = 17 \pm 1$ kPa (red triangles). (right) Peak height h vs. time t for the same experiments. Gray symbols indicate the uncertainty in time by letting $t = 0$ vary by $\pm 0.5/\text{framerate}$.