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Sudden ridge collapse in the stress relaxation of thin crumpled polymer films

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A crumpled thin sheet generally exhibits numerous defects each of which store energy. When subjected to a constant compressive force the height of the crumple decreases logarithmically with time in a process of long-term stress relaxation, which scales over several orders of magnitude—i.e., over time periods from seconds to weeks. We have investigated this scaling behavior for thin polymer films and found that a discontinuous stress relaxation is superimposed on the long-term stress relaxation. The former becomes more pronounced as the polymer sheet thickness increases and is systematically characterized in this study. Effects of ridge length and density are discussed.

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I. INTRODUCTION

When crumpling a thin sheet—for example, a plain piece of paper—defects at varying scale and of different topology are created, which store energy. This is consistent with everyday experience: crumpling a sheet of paper to smaller and smaller size becomes successively harder, even though most of the volume fraction is still taken up by air. In this study we experimentally investigate the long-term behavior of crumpled polymer sheets when subjected to a constant compressive force.

The defects, or singular structures, formed by crumpling a sheet of thickness d, which is much smaller than the lateral size L of the sheet, $d \ll L$, are line defects, called curved ridges [1–3], ending in quasi-point-like defects, called vertices or developable cones [4–7]. An example is depicted in Fig. 1 showing a crumpled polymer sheet [Fig. 1(a)] and a close-up view of ridges ending in a vertex [Fig. 1(b)]. For imaging reasons the otherwise transparent polymer sheet has been spray painted after crumpling. The general geometry of such crumpled sheets has been characterized on several occasions [8–11]. In a process called stress condensation the defects form a connected network with the elastic energy stored mainly in the ridges [12–15]. To find an every day example of such behavior it is sufficient to take a closer look at one's T-shirts when they come out of the washing machine after the spinning cycle. A hopefully less often encountered example is the deformation of the crumple zone during car accidents.

The energy stored in a single ridge was shown to be $E \approx Ydx^{1/3}$ [1–3], where Y is the Young's modulus, d the sheet thickness, and x the length of a ridge. This holds below the regime of plastic deformation and should also be valid for averages of energy and defect length. Matan *et al.* [16] have demonstrated that the size of a crumpled thin sheet, subjected to a constant compressive force, decreases logarithmically with time, according to a relationship

$$h - h_0 = -\beta \log_{10}(t/s), \qquad (1)$$

where $h - h_0$ is the change in height of the crumple and t the time in seconds. For selected materials the scaling behavior of Eq. (1) was shown to hold for up to approximately six orders of magnitude—i.e., for a time period from seconds to several weeks. In this study we show that a discontinuous behavior of sudden ridge collapses is superimposed on the scaling behavior of crumple compression. We further discuss the effects of defect density and ridge length.

II. EXPERIMENTAL METHODOLOGY

Thin polymer sheets of different materials, Melinex-S films (polyester) of varying thickness between 12 and

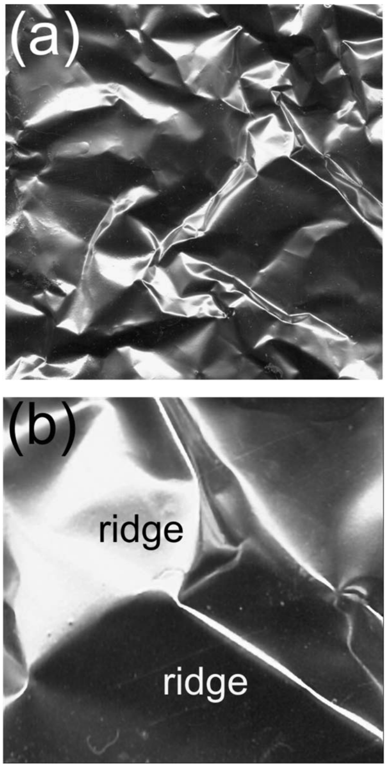

FIG. 1. (a) Topology of a crumpled piece of polymer wrapping film exhibiting a large number of defects (image size 21×21 cm^2). (b) Exemplary illustration of ridges converging into a vertex. The main energy stored in a crumple of a thin sheet is located in the ridges. For illustrative purposes, the polymer film was spray painted before imaging.

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75 μm (Dupont-Teijn Films) and a large-area wrapping film (polyethylene) of thickness 51 μm (surface density 46 gm^{-2}) were crumpled 10 times to approximately the same size before measurement, unless otherwise noted. The reason for this procedure lies in the reproducibility of results and will become apparent from the data reported below. The crumpled sheets were then subjected to a constant force supplied by a total mass of 200 g. This was provided by a metal plate of diameter 15 cm placed horizontally on top of the samples and larger than that of the roughly spherical structures of the crumbled sheets, which have a diameter of approximately 10 cm. The observed changes in height of the crumple are small compared to the diameter of the sample. Lateral dimensions were not subjected to any confinement in order to avoid frictional effects besides the unavoidable friction due to sliding contacts of the sheet material itself. At all times it was assured that the compressive weight was horizontal within a few degrees. Experimental runs which showed an uneven compression were disregarded from the analysis.

Attached to the supporting weight was the low-mass internal part of a linear voltage displacement transducer (LVDT, Solartron DFg5) with a measuring range of 10 mm and sensitivity of 540 mV mm^{-1}. The external part of the LVDT was fixed to an adjustable scaffold in order to ensure optimal working conditions throughout the experiments. Its output was connected to a digital voltmeter (Black Star 4503) with a resolution of 100 μV, and time-resolved data were recorded via a PC. The setup thus provided a nominal resolution for height measurements of the crumpled sheets of approximately ± 0.2 μm. It was calibrated by use of a micrometer screw gauge with resolution of 1 μm over a range of 8 mm, and the very slight deviations from a linear response (0.3%) were accounted for in the analysis by a simple polynomial fit to the recorded calibration data. The resolution of the LVDT also depends on the linearity of the supplied dc driving voltage (10 V), for which a highly stabilized power supply was employed. We estimate the systematic error in the height measurements presented below to approximately $\delta h = \pm 2$ μm, smaller than the symbol size used in the figures. The displayed errors thus refer to the standard deviation or to the reproducibility range of the experimental measurements. The whole apparatus was installed on an optical table to minimize any influences from vibrations, which have been shown to affect the stress relaxation process at large accelerations (2.5 m s^{-2} at 7.5 Hz) [16]. Additionally, the setup was covered by a transparent hood to avoid possible effects of air flow.

The ridge density ρ_{ridges} was determined by scanning respective sheets at a resolution of 1200 dpi, cropping to an area of 10 cm by 10 cm and using digital enhancement via contrast and brightness. The polymer films were coated with reflective gold spray prior to scanning in order to provide adequate contrast and ridge resolution. The number of ridges was determined by manual counting, using the software IMAGETOOL3.0, developed at the University of Texas Health Science Center, San Antonio. A generous error of 10% was assigned to the determination of the number of ridges and thus the ridge density. The average ridge length $\langle L_{\mathrm{ridges}} \rangle$ was determined with IMAGETOOL3.0 after careful calibration from the pixel to millimeter scale. For each length measurement reported, more than 50 individual readings were averaged.

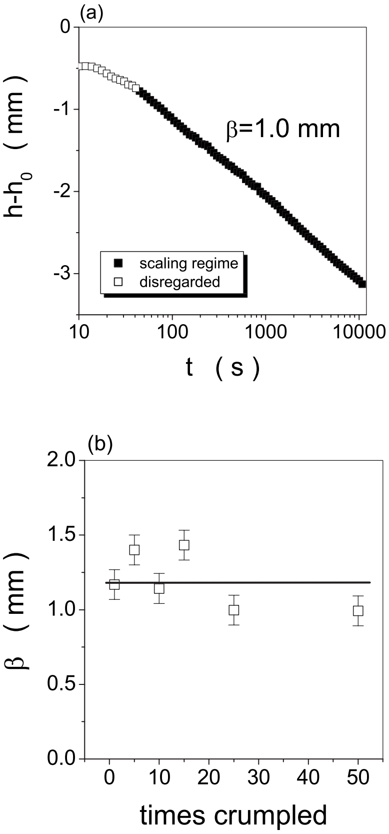

FIG. 2. (a) Exemplary illustration of the scaling relation of Eq. (1) for a standard polymer wrapping film of thickness $d = 51$ μm and surface density $\rho_S = 46$ gm^{-2} over several orders of magnitude in time, corresponding to a time period from seconds to days. (b) Scaling parameter β for experiments after successive processes of crumpling. The fact that β is practically constant illustrates the structural integrity of the polymer foil even after numerous crumpling processes.

III. RESULTS AND DISCUSSION

Figure 2(a) exemplary depicts the scaling behavior of Eq. (1) for a 21×21 cm^2 square sheet of wrapping film of thickness $d = 51$ μm and surface density $\rho_S = 46$ gm^{-2}, which was crumpled for 10 times prior to the measurement. Disregarding the points at the extreme ends of the time regime (open squares), a linear fit to the semilogarithmic data (solid squares) gave a scaling parameter of $\beta = (1.0 \pm 0.2)$mm. This behavior is in general accordance with that reported by Matan *et al.* [16], and scaling according to Eq. (1) is observed for several orders of magnitude in time. It should be noted that all of the experiments were carried out below the regime of plastic deformation, as the latter leads to deviations from the scaling relation of Eq. (1). We have further investigated the reproducibility of the scaling parameter β after successive crumpling events. The results are shown in Fig. 2(b) with a constant scaling parameter β observed with successive crumpling. This illustrates that the structural integrity of

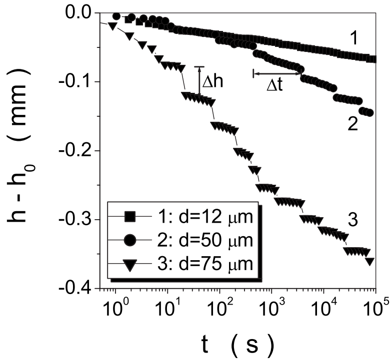

FIG. 3. Long-term stress relaxation behavior for Melinex polymer films of varying thickness d. Discontinuous events of stress relaxation are observed, which are attributed to sudden ridge collapse. This behavior is increasingly pronounced for increasing film thickness.

the polymer films is preserved and evidences the robustness of the results presented below. From an analysis of the (Gaussian) distribution function of β for repeated independent measurements we estimate the reproducibility of the scaling parameter to approximately ± 0.1 mm or 20% for polymer sheets that had been crumpled several times prior to the measurement.

A detailed series of experiments was concerned with the long-term stress relaxation behavior as a function of sheet thickness d. For these investigations commercial polymer films Melinex-S from Dupont-Teijn Films were employed at varying thickness between $d=12$ and 75 μm at a sheet size of 21×21 cm^2. Exemplary measurements of the compression distance $h-h_0$ as a function of time t are depicted in Fig. 3. For thin films ($d=12$ μm, squares) the behavior is very reminiscent of that reported before. But more interestingly, thicker films ($d=50$ μm, up-triangles, and $d=75$ μm, down-triangles) showed clear indications of sudden, discontinuous stress relief, observed as jumps in the $(h-h_0)$ versus $\log_{10}(t/s)$ data, a behavior which has not been reported in previous studies. We believe that the mechanism for this is different from that of jumps observed for laterally vibrated samples [16].

A systematic analysis of this observation is summarized in Fig. 4. The average discontinuous change of height, $\langle \Delta h \rangle$, of the crumple increases linearly with the thickness d of the film, as depicted in Fig. 4(a). It should be noted that $\Delta h(t)$ fluctuates around a mean value, so that it is justified to determine an average discontinuous change of height, $\langle \Delta h \rangle$. The time Δt between successive jumps increases with increasing time t. As time proceeds, the time laps between successive discontinuous events of stress relaxation increases. It is interesting to note that when plotting the latter data $\Delta t(t)$ on a double-logarithmic scale [Fig. 4(b)], data for various different sheet thicknesses all collapse on a single linear curve with a slope being practically equal to unity, $d\Delta t/dt = 1.05 \pm 0.04$. This suggests a more general or universal scaling behavior, which is currently under further investigation. It is presumed that the observed discontinuous stress relaxation events are related to a sudden collapse of ridges of relatively large spatial extension, releasing a comparatively large amount of elastic energy in a very short period of time. Observationally, thick films exhibit a larger number of long ridges as compared to thin films, so a sudden change of crumple height, Δh, is larger for thick films than observed in thin films.

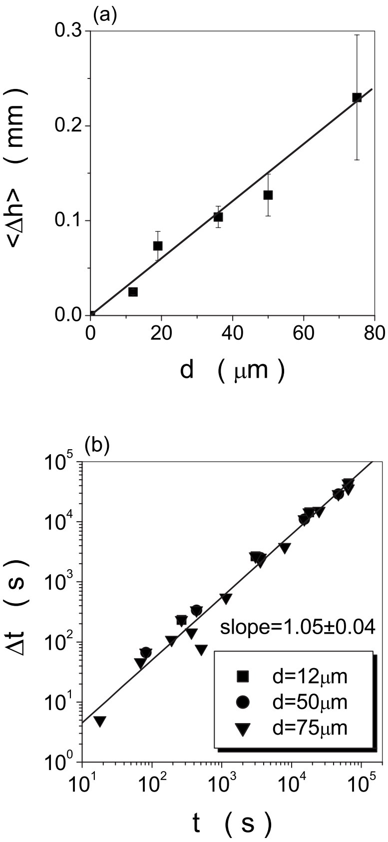

FIG. 4. (a) The average height discontinuity $\langle \Delta h \rangle$ in sudden stress relaxation is increasing with the Melinex film thickness d. (b) The time laps Δt between successive discontinuities increase with time t, being directly proportional on a logarithmic scale, $\Delta t \sim t$, independent of film thickness (solid lines are a guide to the eye).

The experimental results of increasingly larger jumps $\langle \Delta h \rangle$ and longer elapsed time periods Δt between jumps for increasing film thickness suggest that the number of ridges per unit area—i.e., the ridge density ρ_{ridges}—should decrease with increasing film thickness. This is indeed the case as depicted in Fig. 5(a). At the same time the average length of ridges increases according to a square-root power law [Fig. 5(b)].

The scaling parameter β, which characterizes the overall ease of the sheet crumple to further static compression, increases with increasing sheet thickness, as illustrated in Fig. 6(a). A double-logarithmic plot indicates a possible scaling behavior of $\beta \sim d^{1/2}$ within the limits of error, although more detailed investigations are necessary to further validate this relationship. In practical terms the behavior of Fig. 6(a) means that a crumpled structure of a thinner material, but with many more elastic energy storing defects, is more resistant to further compression than a structure of the same size, but of a thicker material with considerably smaller defect density. This is demonstrated in Fig. 6(b), where the scaling

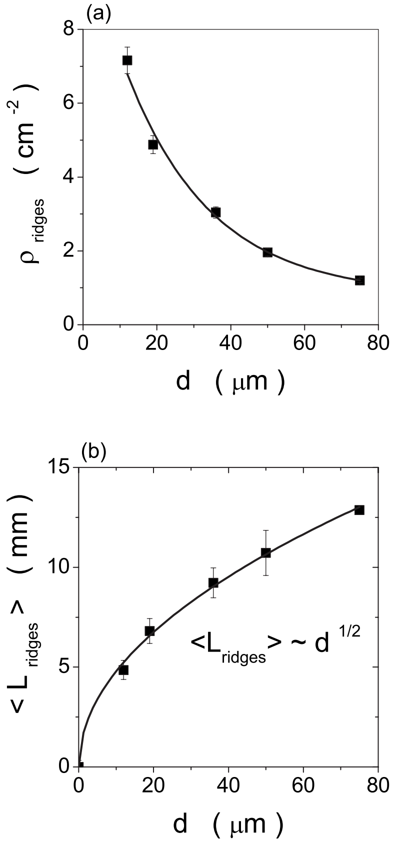

FIG. 5. (a) The ridge density ρ_{ridges} is found to decrease strongly with increasing thickness d of the Melinex polymer films (the solid line is a guide to the eye). (b) At the same time the average length of the ridges, $\langle L_{\mathrm{ridges}}\rangle$, increases with film thickness according to a square-root power law, $\langle L_{\mathrm{ridges}}\rangle \sim d^{1/2}$. The solid line is a fit to the latter relationship.

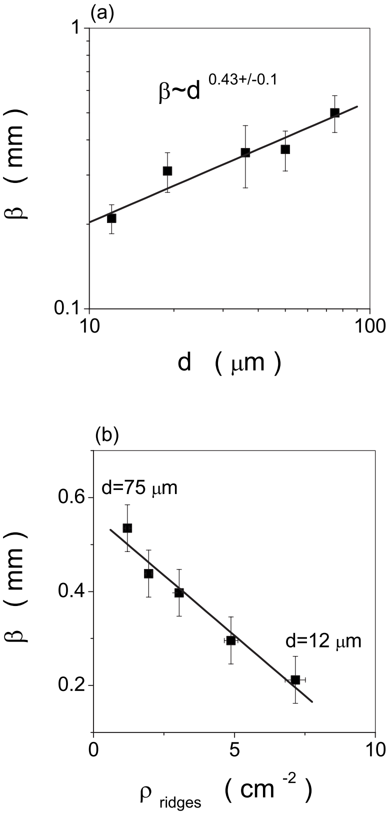

FIG. 6. (a) The scaling parameter β of the Melinex films increases with film thickness d. A double-logarithmic representation suggests an increase according to $\beta \sim d^{1/2}$ within experimental errors. (b) At the same time β decreases with increasing ridge density ρ_{ridges}, parametrized by film thickness as indicated (solid line is a guide to the eye). This behavior is due to the fact that many more ridges of much smaller dimension are formed in thin films, while thick films only exhibit a small number of ridges of larger length.

parameter β is depicted as a function of the ridge density ρ_{ridges}, parametrized by the sheet thickness d as indicated.

IV. CONCLUSIONS

We have experimentally characterized the scaling behavior of crumpled thin polymer sheets during the process of long-term stress relaxation. The scaling parameter β, which describes the ease to further compression of the crumple, is practically constant for successive crumpling due to the structural integrity of the polymer film material being preserved. An increasing number of ridges of smaller size are observed during successive crumpling processes. The scaling parameter also increases with increasing sheet thickness. This is due to a substantial decrease of the ridge density with increasing sheet thickness. Crumple compression is thus easier for thicker materials with fewer, but longer ridge defects. The stress relaxation process was found to be superimposed by discontinuities, which are attributed to sudden ridge collapse. This behavior is increasingly apparent for thicker films and was investigated in detail. The sudden change of crumple height increases linearly with increasing polymer film thickness. The time period between successive ridge collapses increases with elapsed time. Interestingly, in a double-logarithmic plot all data collapse, indicating a universal relationship of $\Delta t \sim t$.

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