Supporting Information

Highly deformable, ultra-thin large-area PMMA films

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Figure S1 shows a typical image of a scratch introduced on the PMMA film, cast from 3 wt% solution, in order to measure its thickness by AFM. The ridges formed along the edge of the scratch are due to the polymer which is displaced during scratching. To improve accuracy in thickness measurement, the average depth of each scratch is measured (Figure S1).

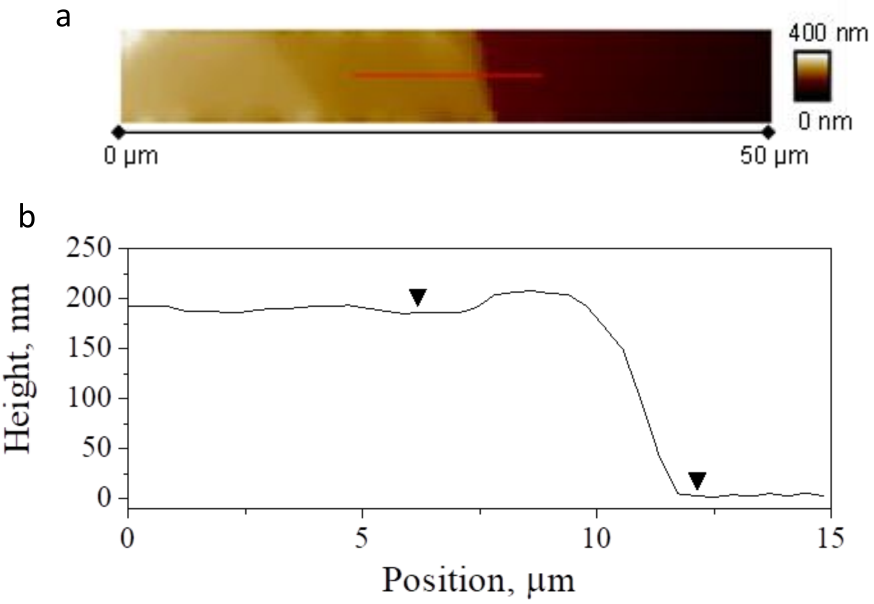

Figure S1. *Image of the scratch (a) and its cross-section (b) on a PMMA film cast from 3 wt% solution.*

In the reported tensile tests, PMMA specimens were deposited on silicon slices provided with a notch on both the bottom and the top surface. The pre-notch on top provided a window where the specimen was free-stranding (i.e., specimen testing area), while elsewhere it was in contact with the substrate (i.e., specimen clamped region). As previously reported in the literature, the interaction of a sub-micrometer thick film with silicon is usually strong enough to provide secure clamping during a tensile test [1–3]. In order to investigate the capability of the interaction with silicon to prevent our PMMA films from sliding, we performed the following experiment. A 250 nm thick PMMA film was deposited on a silicon slice, which was then engraved in order to develop a tiny fracture line on top, as shown in Figure S2[1].On the right of the sample area as shown in Figure S2, there was a completely freestanding PMMA region spanning a window of about 120 µm (not shown in Figure S2). Direct observation of the sample revealed the presence of optical fringes on a region of the film crossing the fracture line; such fringes can be considered an indication that the film partially detached from the substrate itself, as a consequence of the fracture process; but, elsewhere PMMA was strongly interacting with silicon and did not show any sliding under the application of a uniaxial load (along the horizontal direction in Figure S2a-c) until failure. Such conclusion can be drawn by tracking the displacement of two regions, R1 (the black squared region in Figure S2; not showing optical fringes) and R2 (the red squared region in Figure S2; showing optical fringes), of the PMMA film with respect to the underlying substrate (which was connected to a load sensor). The application of an increasing load caused the stretching of the freestanding PMMA film (not shown in Figure S2) and the opening of the initial fracture line in the silicon slice, as shown in Figure S2a-c; the opening of the fracture linethen caused the PMMA film across the fracture line to detach and slide with respect to the underlying silicon substrate (red markers in Figure S2d). On the contrary, where no fringes appeared, PMMA did not significantly move with respect to the substrate (black markers in Figure S2d).

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[1] This procedure differs from the preparation carried out before tensile tests, as in these latter cases the fracture line induced in the silicon slice reaches the top pre-notch and, thus, results to be more than ten micrometers off from the PMMA film.

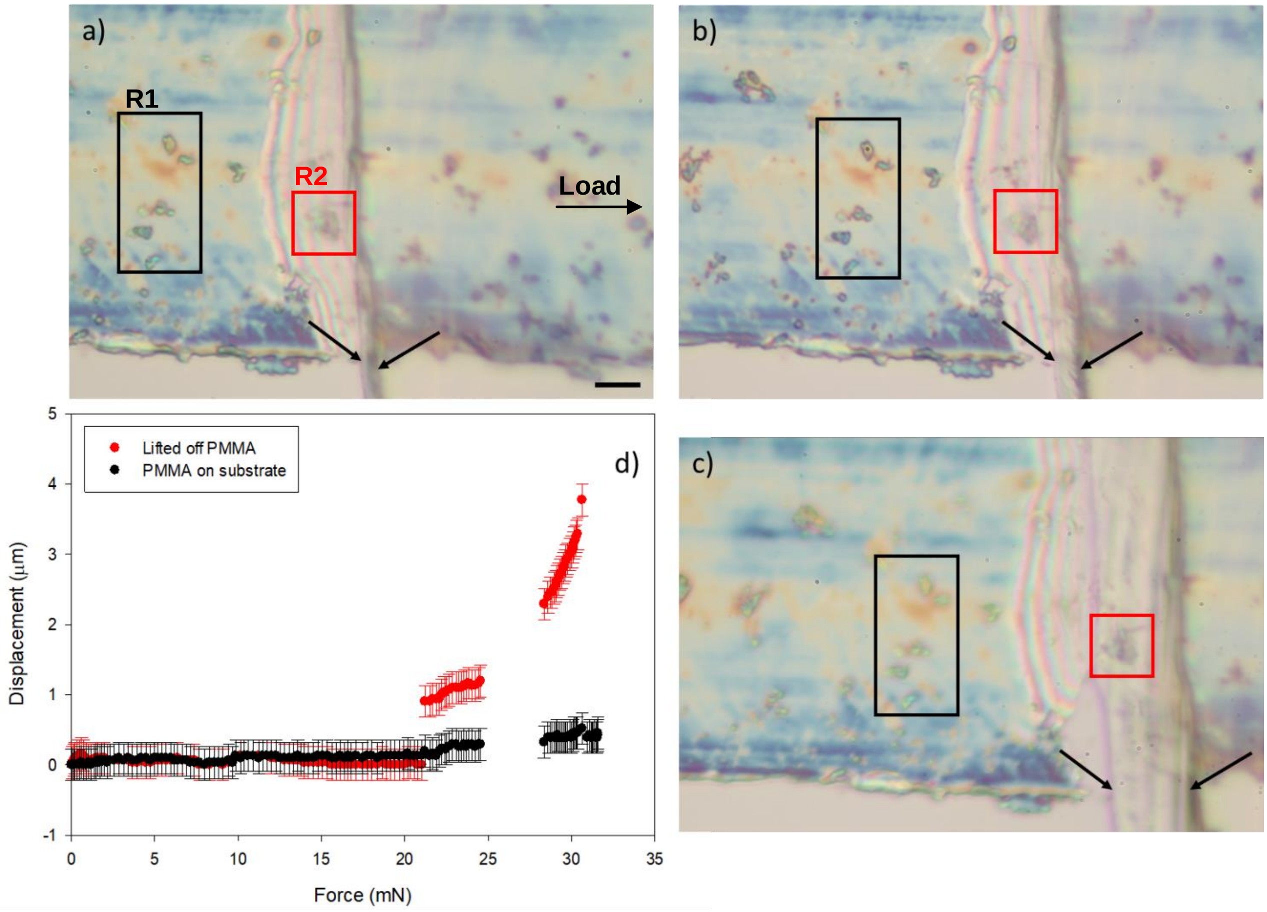

Figure S2: *a) A 250 nm thick PMMA film deposited onto a silicon slice (connected to a load sensor on the left and subjected to a uniaxial horizontal force to the right), which was then fractured (the few micrometers wide fracture line is indicated by the arrows). The PMMA region crossing the fracture line shows optical fringes, which indicate that the film is partially detached from the substrate. When the right side of the silicon slice moves under the load (b-c), the film region R1 moves rigidly together with the underlying silicon slice, while the region R2 does not. d) Relative displacement of reference points belonging to region R1 and R2 with respect to the underlying silicon substrate. Reference points of region R1 do not show significant displacement. On the contrary, reference points of region R2 move together with the underlying silicon up to about 20 mN, then complete detachment and failure occur. The load reported in the graph was applied to a freestanding PMMA film of about 3 mm width located on the right of the area focused in panels a-c. Scale bar: 5 µm*

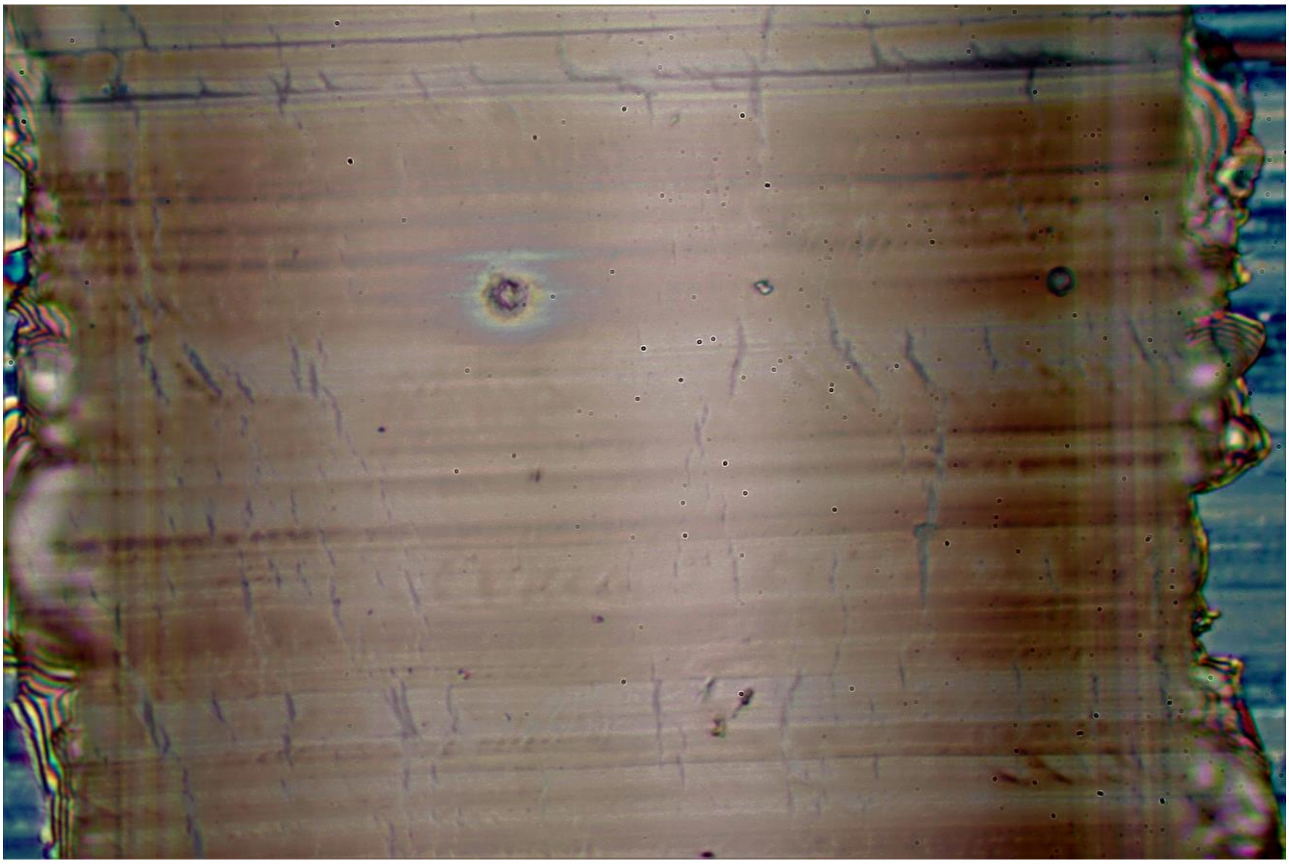

Figure S3:*A 200-nm thick PMMA film at 6% strain.*

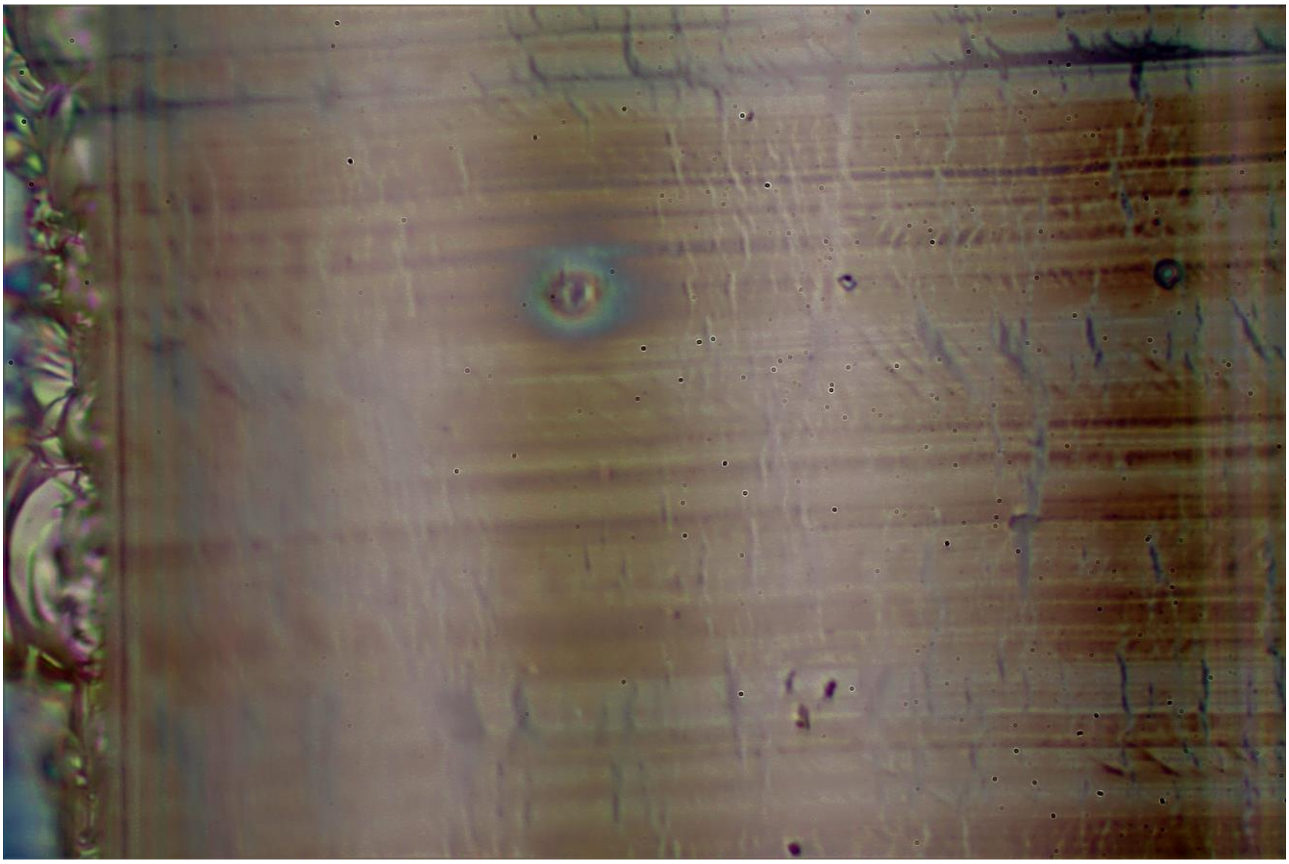

Figure S4: *A 200-nm thick PMMA film at 17% strain.*

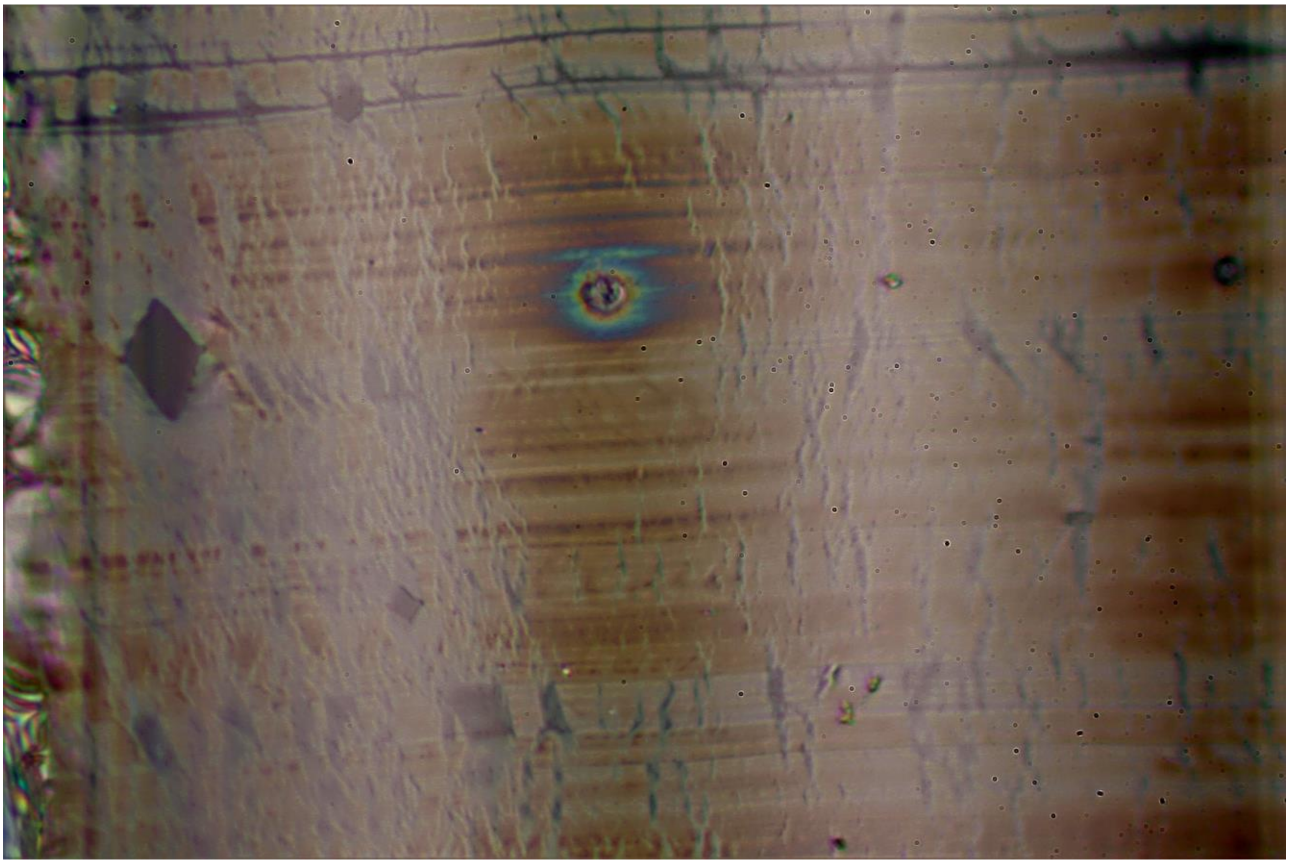

***Figure S5**: A 200-nm thick PMMA film at 25% strain.*

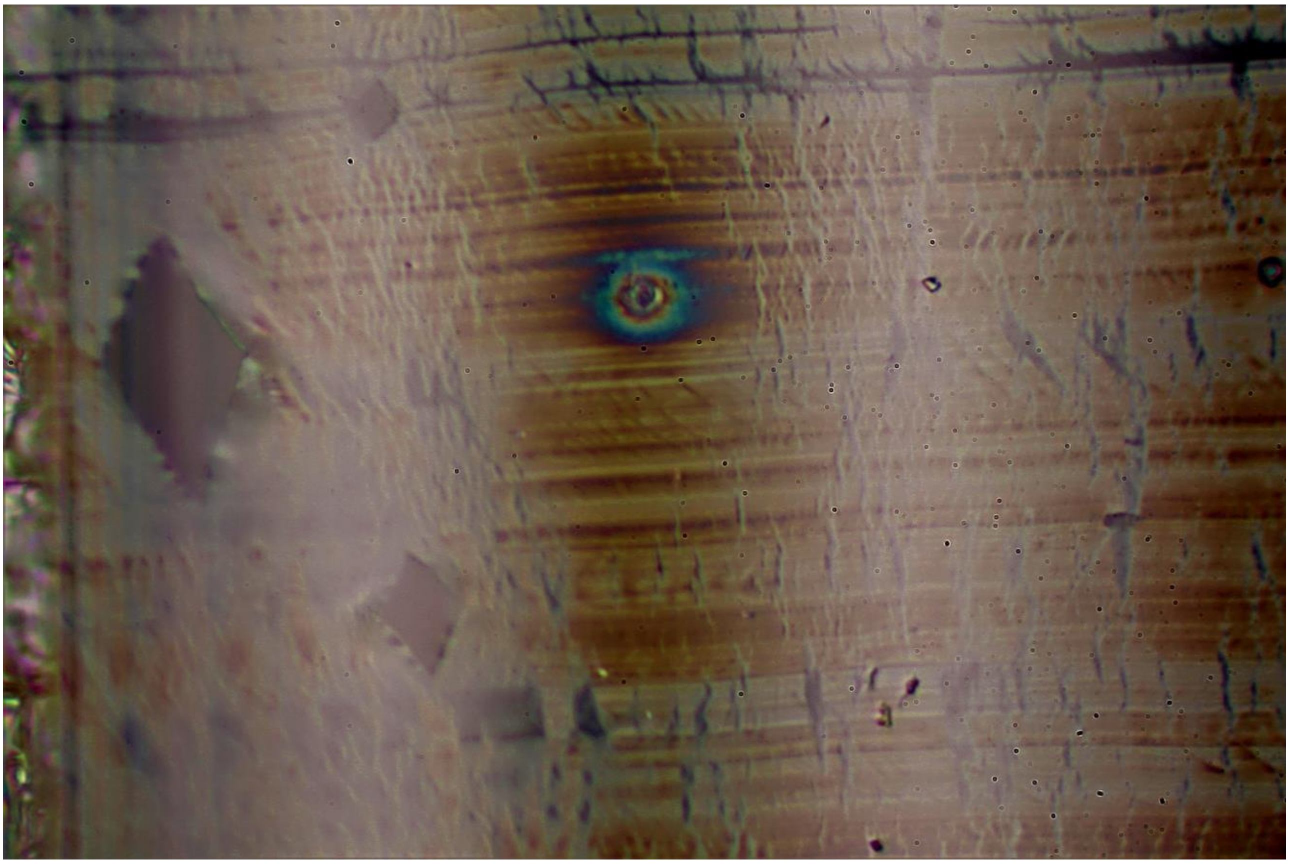

***Figure S6**: A 200-nm thick PMMA film at 30% strain.*

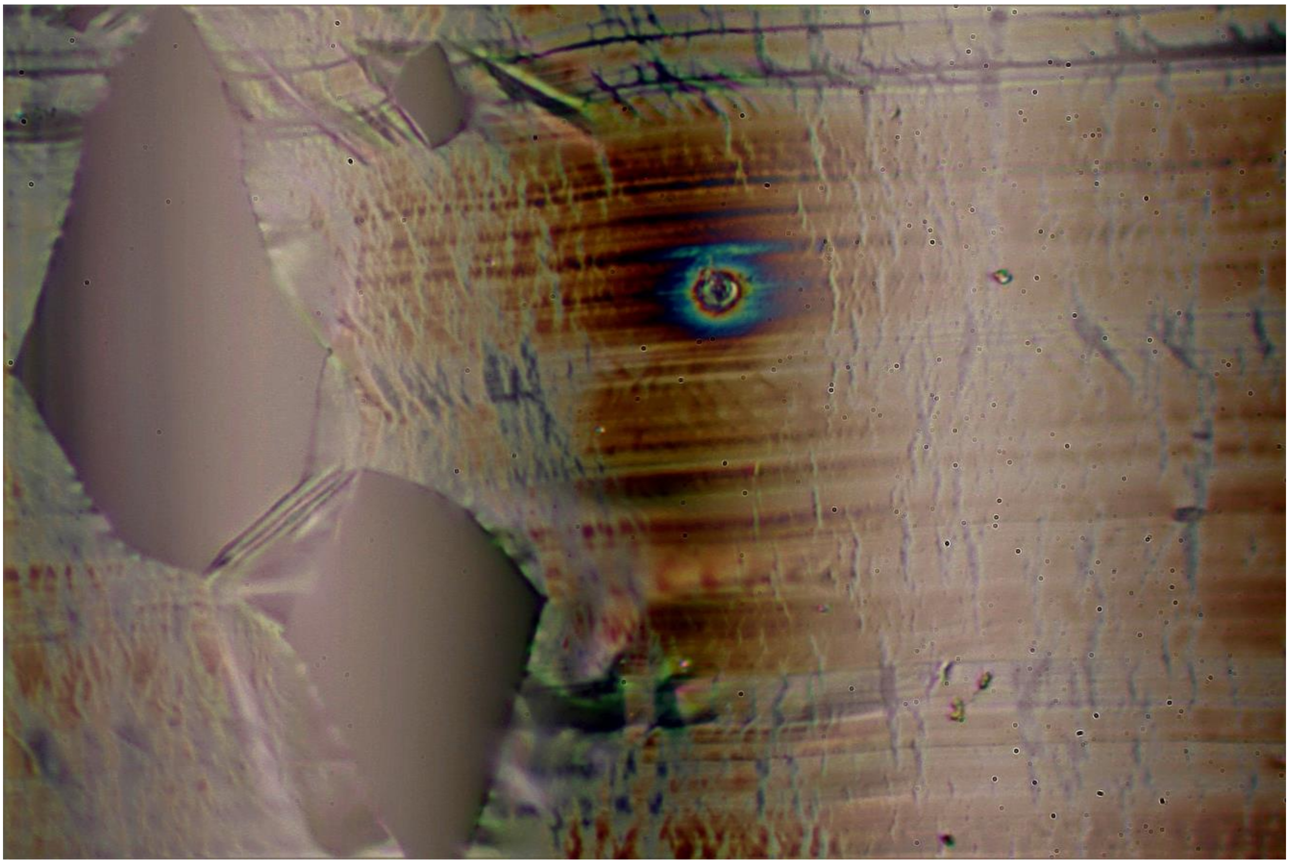

Figure S7: *A200-nm thick PMMA film at 43% strain.*

Supporting references

(1) Cao, C.; Zhang, Z.; Amirmaleki, M.; Tam, J.; Dou, W.; Filleter, T.; Sun, Y. Local Strain Mapping of GO Nanosheets under in Situ TEM Tensile Testing. *Appl. Mater. Today***2019**, *14*, 102–107.

(2) Cao, C.; Mukherjee, S.; Howe, J. Y.; Perovic, D. D.; Sun, Y.; Singh, C. V.; Filleter, T. Nonlinear Fracture Toughness Measurement and Crack Propagation Resistance of Functionalized Graphene Multilayers. *Sci. Adv.***2018**, *4* (4), eaao7202.

(3) Frankberg, E. J.; Kalikka, J.; Ferré, F. G.; Joly-Pottuz, L.; Salminen, T.; Hintikka, J.; Hokka, M.; Koneti, S.; Douillard, T.; Le Saint, B.; Kreiml, P.; Cordill, M. J.; Epicier, T.; Stauffer, D.; Vanazzi, M.; Roiban, L.; Akola, J.; Fonzo, F. Di; Levänen, E.; Masenelli-Varlot, K. Highly Ductile Amorphous Oxide at Room Temperature and High Strain Rate. *Science***2019**, *366* (6467), 864–869.