FIGURE S1 (a) Optical image of typical line series for analysis with $z$ focus increasing from bottom-to-top and power increasing from right-to-left. The region of best focus for AFM analysis is identified by locating where the lines are visible at low power (marked for this image by the white dashed line). (b) AFM image of region of best focus with $z$-scale of 150 nm.
FIGURE S2 Evolution of height profiles for PS films with various MW for film thicknesses (a-c) ~50 nm and (d-f) ~110 nm: (a,d) the maximum profile height ($h_f$) versus power, (b,e) the combined profile FWHM ($w_f$) vs. power, and (c,f) normalized profile shapes at the highest measured power.
FIGURE S3 Averaged AFM phase line scans of single dewetted lines. Large deviations correspond to changes in height while the level regions correspond to the exposed substrate (center) and the surrounding film (far left and right of the plot). In the case of the bare silicon sample (black trace), there is a difference in phase value between the substrate and surrounding film, while there is no difference in the experiment with the PS brush, indicating that the surface retains the brush component despite the high temperatures briefly encountered during the exposure.

FIGURE S4 Results of temporal simulation of complete dewetting of a 50 nm PS film, showing the (a) height \( h \), (b) temperature \( T \), (c) height change rate \( \frac{dh}{dt} \), and (d) thermal gradient magnitude \( |\nabla T| \) profiles. It can be seen that the thermal profile is off-center and the thermal gradient is strongest towards the advancing front due to the ARC effects. This results in the height change occurring mainly in advance of the laser spot. \( h \)-scale in (a) is 0-160 nm, \( T \)-scale in (b) is 50-500 °C, \( \frac{dh}{dt} \)-scale in (c) is -2.4-0.8 μm/s, and \( |\nabla T| \)-scale in (d) is 0-100 K/μm.
Figure S5 Proposed mechanism for evolution of oscillating lines based on prior research on bilayer films.\textsuperscript{1,2} (a) The initial state of the film is a high MW PVP layer placed above a low MW PS layer. When the laser initiates dewetting, the more viscous PVP top layer displaces the PS bottom layer creating a region of the film of higher PVP concentration and therefore higher molecular weight (b).
FIGURE S6 AFM image of a PVP/PS bilayer sample written with alternating-scan direction lines. A large initial feature is always observed at the start of the line corresponding to the additional dwell that occurs as the stage ramps to full speed. Color scale is 120 nm.

MOVIE S1 Simulated time evolution of film height profile during writing at 95 mW of absorbed power. At this power the film is not completely dewetted, but a steady state height profile is rapidly achieved with a majority of displaced polymer being pushed ahead of the moving beam.

MOVIE S2 Simulated time evolution of the temperature profile in the film shown in MOVIE S2.

REFERENCES