Electronic Supplementary Material (ESI) for Soft Matter.

Observation of dynamical heterogeneities and their time-evolution on the surface of an amorphous polymer

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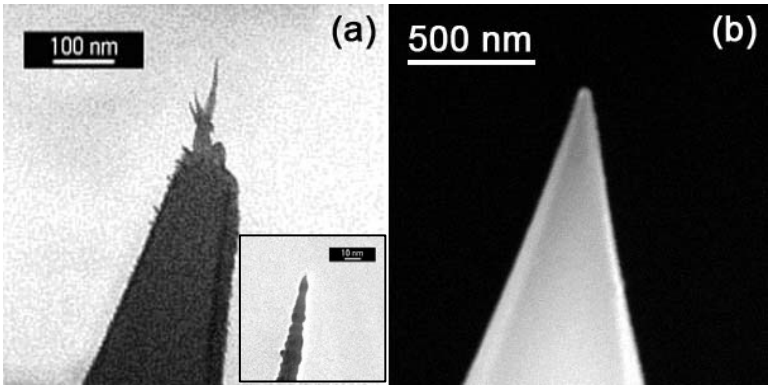

Fig. S1. (a) Morphology of the diamond-like tip with a ~1-nm apex used in the current work. The insert is a TEM image of the sharp part of the tip. (b) Morphology of a conventional AFM tip.

Calculation of the degree of HDs similarity upon time-evolution. By repeatedly scanning a small area on the surface of PS films, the time-evolution of heterogeneous domains (HDs) can be observed. In addition, a characteristic lifetime of a portion containing of tens of HDs can be estimated by using normalized cross correlation (NCC) analysis of this portion with scanning time (1). The formula of NCC, denoted as $C(u, v)$, is presented in equation (3) of the main text. For charactering the behavior of $C(u, v)$ on the phase shift image with the scanning time, we divided each phase shift image into 8 equal patterns of 64 × 512 pixel starting from the turning position, denoted as 1 to 8 on the first scanning image and 1′ to 8′ on the second scanning image, as shown in Fig. S2. Then, each two corresponding parts on two images (Fig. S1a and S1b) were used to calculate the average value $\langle C(u, v)\rangle$ over the whole area. For example to calculate $\langle C(u, v)\rangle$ between pattern 1 and 1′, an arbitrary portion of 25 × 50 pixel, i.e. ~ 50 HDs, centered at a position (x_0, y_0) in the pattern 1 was selected. The NCC of this portion was calculated over the pattern 1′. The maximum value of $C(u, v)$ was obtained indicating that the selected portion shifted to a new position $(x_0 - u, y_0 - v)$ in the pattern 1′. Such a calculation was performed for all portions of 25 × 50 pixel centered at different positions in the pattern 1 to provide an average value $\langle C(u, v)\rangle$. Because it took 128 s to complete the scanning of the patterns 1 and 1′, the average value $\langle C(u, v)\rangle$ calculated like this represents the degree of the similarity of tens of HDs

after 128 seconds of scanning. Repeating this procedure provides us the average value <$C(u, v)$> of tens of HDs in all eight patterns, i.e. at different scanning times, which is shown in Fig. 3 of the main text.

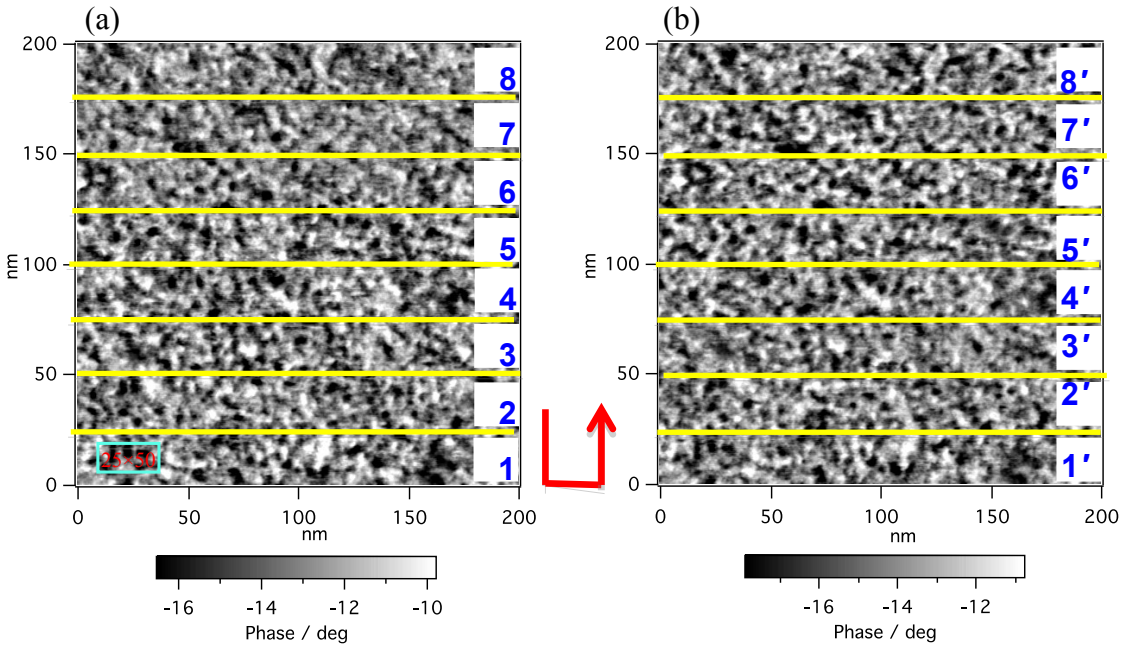

Fig. S2. Phase shift images obtained on the same region of the neat 97-nm thick PS film at the first (a) and second (b) scanning. Each image is divided into 8 equal patterns of 64 × 512 pixel for estimating the degree of similarity between two corresponding patterns. Small turquoise box represents a portion of 25 × 50 pixel containing ~ 50 HDs. All such portions centered at different positions are used to calculate the average value <$C(u, v)$> over each pattern.

1. D.-M. Tsai, C.-T. Lin, J.-F. Chen, *Pattern Recognit. Lett.*, 2003, **24**, 2525-2535.