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Supplementary Information

Revealing the detailed structure in flow-induced crystallization of semicrystalline polymers

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(1) Here we will discuss the influence of the polydispersity on the scattering pattern at small strains.

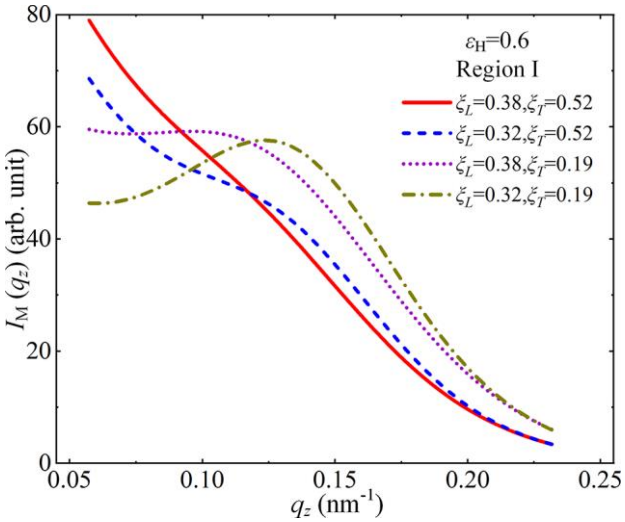

Figure S1. Calculated meridian scattering intensity profiles, $I_{\mathrm{M}}(q_z)$. The red solid line gives the result at $\varepsilon_{\mathrm{H}} = 0.6$ in region I ($\xi_L = 0.38$ and $\xi_T = 0.52$). Moreover, we plot three other cases for comparison. The blue dashed line represents the result for $\xi_L = 0.32$ and $\xi_T = 0.52$, the purple dotted line represents the result for $\xi_L = 0.38$ and $\xi_T = 0.19$, and the brown dash-dot line represents the result for $\xi_L = 0.32$ and $\xi_T = 0.19$. One can find that by decreasing ξ_L or ξ_T, the scattering peak appears.

It is seen that in region I, the scattering peak of the meridian axis is absent. As shown below, this is due to the large polydispersity at low strains. In Figure S1, we show the calculated meridian intensity for $\varepsilon_{\mathrm{H}} = 0.6$ in region I (red solid line). Here we have $\xi_L = 0.38$ and $\xi_T = 0.52$. Moreover, we also calculate three other cases. The blue dashed line represents the result for $\xi_L = 0.32$ and $\xi_T = 0.52$, the purple dotted line represents the result for $\xi_L = 0.38$ and $\xi_T = 0.19$, and the brown dash-dot line represents the result for $\xi_L = 0.32$ and $\xi_T = 0.19$. In these three additional cases, the only different parameters from those at $\varepsilon_{\mathrm{H}} = 0.6$ are ξ_L and ξ_T. In fact, $\xi_L = 0.32$ and $\xi_T = 0.19$ are typical results at $\varepsilon_{\mathrm{H}} = 0.9$ in region II. It can be found that, for the configuration at $\varepsilon_{\mathrm{H}} = 0.6$ (region I), due to the large polydispersity of L, the scattering peak disappears. In addition, the large polydispersity of T also enhances the low-q upturn.

(2) In this part we will show that by assuming that the long period (L) or the kebab thickness (T) has a bimodal distribution, it is not able to fit $I_2^0(q)$ and $I_4^0(q)$ simultaneously.

Figure S2 give the fits by assuming L or T has a bimodal distribution in the heterogeneity manner. It is clear that none of them lead to satisfactory fits.

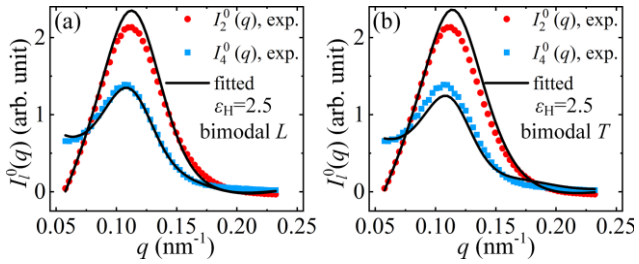

Figure S2. The data fits of $I_2^0(q)$ and $I_4^0(q)$ for XL-HDPE after isothermal crystallization at $\varepsilon_H = 2.5$ in region IV assuming that (a) the long period L or (b) the kebab thickness T has a heterogeneous bimodal distribution, respectively.

(3) In this part, we will show our preliminary result on tracking the formation and growth of kebabs with the SHE method.

As we shown in the main text, the term $I_2^0(q)$ is consistent with the geometry of external deformation, and reflects the major characteristics of the structure. Figure S3(a) shows the evolution of $I_2^0(q)$ at $\varepsilon_H = 2.3$ during the crystallization process. One can get a qualitative impression about the formation and growth of kebab. At the beginning, the $I_2^0(q)$ is negative, which shows that the intensity of shish dominates the spectrum. Then $I_2^0(q)$ becomes positive, with a peak at about q = 0.12 nm^{-1} due to the period of kebab. This indicates the formation of kebabs. $I_2^0(q)$ keeps increasing until t ~ 1860 s, and then gradually stabilizes.

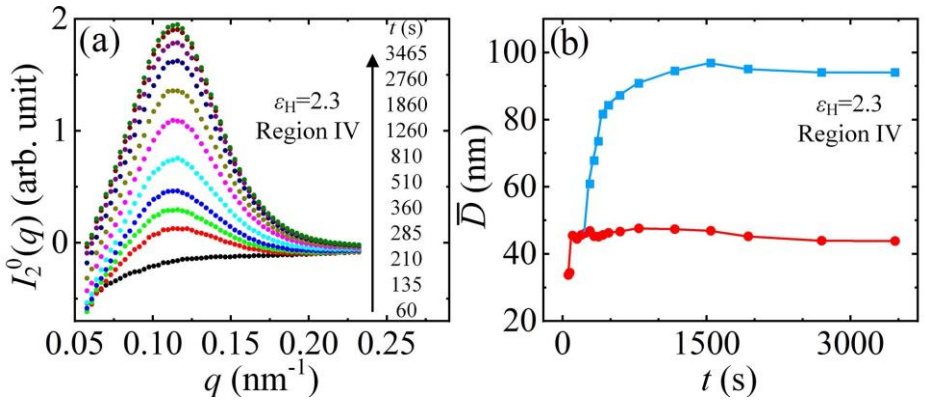

Figure S3. (a) The evolution of $I_2^0(q)$ with time at $\varepsilon_H = 2.3$ during the crystallization process. (b) Kebab diameter as a function of time at $\varepsilon_H = 2.3$.

To quantitatively track the formation of kebab, we fit the first few anisotropic terms in SHE with the shish-kebab model given in the main text at different times at $\varepsilon_H = 2.3$ (region IV), and the fitted average diameter of kebab as a function of time is shown in Fig. S3(b). As one can see, the growth of the kebab exhibits a kinetic heterogeneity, in the sense that at t_c ~ 300 s the bimodal distribution of kebab diameter appears. Before t_c, the unimodal distribution can lead to a very good fit. While after t_c, one must assume a bimodal distribution of kebab diameter to fit the spectra. According to this result, we conjecture that there are differences in the end time of lamella growth in different spatial regions, which may be related to the number of crystallizable chains at the growth fronts of lamellae. We emphasize that further investigations are needed to reveal the mechanism of this phenomenon.