Supplementary Information to "Crystallization of semiflexible polymers in melts and solutions"

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Figure S 1: (a) Comparison of valence (on bonds between two monomers adjacent along the chain) and non-valence (between non-bonded monomer units) potentials. Legend: (1) flexible bond potential (case of phantom chains), (2) stiff bond potential (case of non-phantom chains), (3) repulsion potential of volume interactions for the case of phantom chains (with cutoff radius 1.0), (4) repulsion potential of volume interactions for the case of non-phantom chains (with cutoff radius 1.0). Both bond potentials are defined by the expression $U = k(l - l_0)^2/2$. In the case of phantom chains, the values of parameters are k = 4 and $l_0 = 0$, while in the case of non-phantom chains, which we have used in our simulations, the values of parameters are k = 150 and $l_0 = 0.2$; this allows us to include in the model the intrachain stiffness and to avoid chain crossing (non-phantom chains). Two soft repulsion potentials are defined by the equation $U = a_{ii}(1-r)^2/2$, where the parameter value for phantom chains is $a_{ii} = 25$, and in our case we used $a_{ii} = 150$ to keep the chains to be non-phantom. The interval [0, 1.1] is shown for better visibility. (b) Normalized histograms of distances between bonded (blue line) and non-bonded (pink line) monomer units confirm the absence of self-intersections in the course of simulations. Values for non-bonded distances have been already multiplied by $\sqrt{2}$ for direct comparison with the nonphantomness condition $\sqrt{2}r_{min} > l_{max}$ (see the text). The data are obtained for the final conformation of the system with polymer volume fraction $\varphi = 0.2$.



Figure S 2: The radial distribution function for systems with different polymer volume fraction: (a) $\varphi = 0.2$, (b) $\varphi = 0.5$, (c) $\varphi = 1$. The curves were calculated for the whole systems (polymer and solvent) as well as for amorphous and crystalline domains separately (see the legend). The first maximum at $r \approx 0.5$ comes from the neighbors along the chain.



Figure S 3: Steinhardt local bond order parameters Q6 vs. Q4 (a,c) and Q6 vs. W6 (b,d) for amorphous and crystalline phases (indicated in the legend) for the systems with $\varphi = 0.2$ (a,b) and $\varphi = 1$ (c,d). For each system, the final conformations were used.



Figure S 4: Distributions (box plots) of eigenvalues S_1 , S_2 , S_3 of orientational tensor $Q_{\alpha\beta}$ for systems with different polymer volume fraction (indicated on the top of each graph). Crystalline and amorphous phases are considered separately, while in the crystalline phase each crystallite having the size larger than 10 beads is analysed separately (data for small crystallites were not taken into account). Orange line is the median, and the blue point is the average value weighted with the crystallite size (its numerical value is also shown at the top of the graph). Boxes include the second and the third quartiles, and circles indicate outliers. For each system, the final conformations of 5 independent runs were used.



Figure S 5: Dependencies of eigenvalues S_1 , S_2 , S_3 of orientational tensor $Q_{\alpha\beta}$ on the size of crystalline clusters for systems with different polymer volume fraction (indicated on the top of each graph). The data for amorphous phase are not shown here. For each system, the final conformations of 5 independent runs were used.



Figure S 6: Distributions (box plots) of parameter ψ_6 , which indicates 2D hexagonal packing in the plane perpendicular to the director of a crystallite, for different polymer volume fractions (indicated on the top of each graph). Data for crystalline and amorphous phases are represented separately, while in the crystalline phase each crystallite having the size larger than 10 beads is analysed separately (data for small crystallites were not taken into account). Orange line is the median, and the blue point is the average value weighted with the crystallite size (its numerical value is also shown at the top of the graph). Boxes include the second and the third quartiles, and circles indicate outliers. For each system, the final conformations of 5 independent runs were used.



Figure S 7: Dependencies of ψ_6 on crystallite size for different polymer volume fractions (indicated on the top of each graph). For each system, the final conformations of 5 independent runs were used.



Figure S 8: Dependencies of ψ_6 on the largest eigenvalue S_1 of orientational tensor. Points near [0,0] correspond to the amorphous phase.



Figure S 9: The entanglement length N_e depends quite strongly on the polymer volume fraction φ but depends very weakly on time (after 100 DPD steps). The calculation was carried out in a similar manner to the procedure developed in M. Kröger's group. This indicates that the entanglement length stays almost unchanged while the initial crystallite seeds are formed and crystallization process goes on. These data were obtained in simulation runs without the initial pre-structuring for 10⁶ DPD steps in θ -solvent.



Figure S 10: Movie snapshot. Original mp4 file is attached. It consists of 1000 frames at 30 fps rate. The movie shows first 10^6 DPD steps of simulation of crystallization process in solution with polymer volume fraction $\varphi = 0.2$ without preliminary structuring in θ -solvent. Two chains are colored using the rainbow color gradient in order to make local changes during the crystallization process better visible, other polymer chains are colored gray semitransparent. Solvent particles are not shown in the movie. The bonds between chain beads are not shown in the movie.



Figure S 11: Degree of crystallinity along the chain for the systems with polymer volume fraction $\varphi = 0.5$ (a) and $\varphi = 1$ (b). Gray lines correspond to the 5 independent runs, and the red curve is their mean.