

## Transferability of the Coarse-Grained potentials for *trans*-1,4

### Polybutadiene

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To confirm the equilibration of our simulated systems or to ensure that the resulting lamellar crystal structure at 300 K is thermodynamically stable, we have undertaken three independent simulations for each model system. Here we take our CGMD results obtained from CG model without CG torsion potentials as an example. We note that the polymer crystallization from the isotropic melt to the lamellar structures occur in all three samples. Figure S1 show the time-evolution of the intermolecular LJ energy and density in the three independent CG simulations at 300 K.

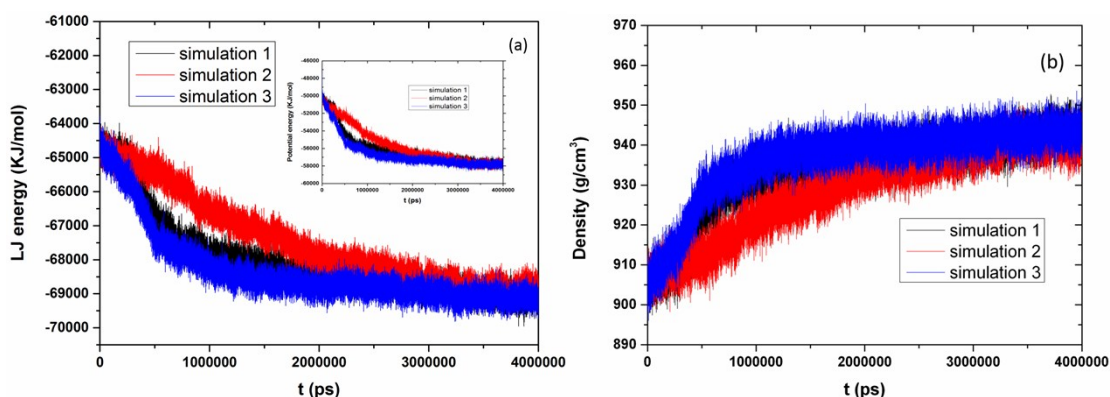


Fig. S1. (a) Evolution of the intermolecular LJ energy in three independent CG simulations of *trans*-1,4-polybutadiene at 300 K. Insert: the time evolution of the system potential energy. (b) Evolution of the system density in three independent CG simulations of *trans*-1,4-polybutadiene at 300 K.

Because these three CGMD simulations are started by quenching the amorphous melt at 413 K to 300 K, the intermolecular LJ energy shows a dramatic decrease at first. After 2000 ns the decrease of this LJ energy becomes more slowly and until 3500 ns it appears to reach a steady state i.e., it varies by less than 0.8% over the last 500ns. Additionally, as indicated by the insert plot, the instantaneous system potential energy change follows this LJ energy profile and the intermolecular LJ energy is the primary contributor to the large change in potential energy. The decrease in the intermolecular LJ energy is indicative of a closer packing between monomers on separate chains, suggesting the polymer chains order into lower energy structures. Meanwhile, the

bulk density in three samples, after an initial sudden rise due to the quenching effect, grows gradually and finally reaching a plateau at about  $0.943\text{g/cm}^3$ , which supports the observed polymer crystallization with the first-order liquid–solid transition character. Moreover, the time evolution of density is synchronized with the (intermolecular LJ) potential energy. Comparison of these three parallel simulations shows no significant variation in the final results. Indeed, these essentially identical results provide further evidence that the equilibration times we adopted at 300 K is sufficient to obtain stable density and constant potential energy and our simulated systems are equilibrated. This further validate that the chain-folded lamellae is an equilibrium structure not a metastable state.

To provide more insight on the agreement between the crystallization structures obtained from atomistic and CG simulations, we followed Lacey's work and calculated the (local) degree of order of the chain. The resulting degree of order averaged over last 100 ns trajectories from MD simulations on both atomistic model and CG model with torsion potentials is presented in Fig. S2.

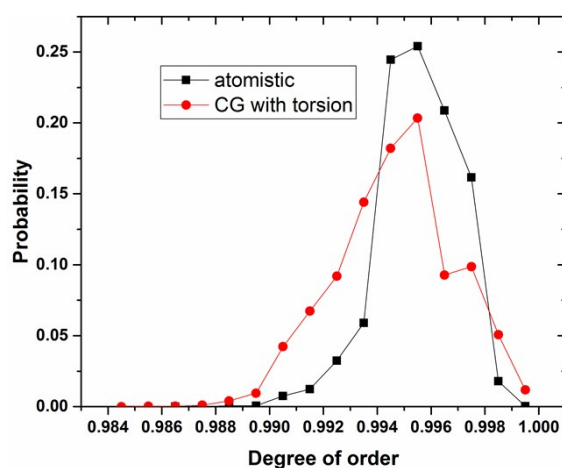


Fig. S2. The distribution of local orientational order degree of individual chain averaged over last 100 ns trajectories from MD simulations on both atomistic model and CG model with torsions.

We see that both distribution profiles have similar shapes and according to Lacey's criterion the (local) degree of order in both atomistic and CG model systems at 300K is very high, suggesting a significant orientational ordering of chains in lamellae. Moreover the result from atomistic model system is a little higher than that from CG one, which is consistent with the tendency of crystallinity parameter, indicating that two observables which by definition respectively measure the global orientational order parameter of a poly-crystalline system or the local orientational order parameter of the chain can be used alternatively to characterize the internal orientation structure of ordered domains.

Besides comparing the local and the global orientational order degree of atomistic and CG model systems, we also undertake the system structure comparisons by examining RDF (which is presented in the main text) and static structure factor (see Fig.S3 below) of *trans*-1,4-PB system at 300 K. In such a plot,  $S(q)$  can be separated into three regimes. The first one is the high  $q$  regime,  $q > 2\pi/(\text{CG bond-length}) \approx 14 \text{ nm}^{-1}$ , which corresponds to the small length scale. The low  $q$  regime of  $q < 2\pi/(\text{radius of gyration}) \approx 1 \text{ nm}^{-1}$  corresponds to the overall chain dimensions. In-between is the intermediate range of  $1 \text{ nm}^{-1} < q < 14 \text{ nm}^{-1}$ .

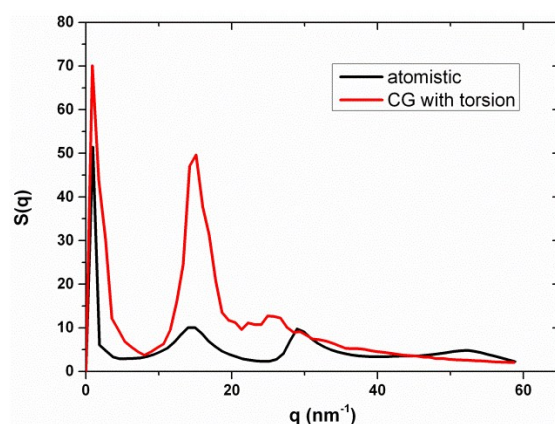


Fig. S3. The static structure factors of the final state in atomistic simulation and CG simulation with torsions.

From the figure we can see that a significant sharp peak at low values of  $q$  appears in both  $S(q)$ , indicating the existence of a global long-range ordering in both atomistic model system and CG model system with torsions, consistent with the snapshot visualization that lamellae has developed in the lower temperature liquid. Additionally, the CG  $S(q)$  has almost the same shape as that of its counterpart atomistic model system except the magnitudes of the peaks becoming relatively large. We note that the box dimensions in atomistic simulation and CG simulation with CG torsion potentials are not completely consistent due to the use of NPT ensemble. So the resolution of  $q$  in two simulations is a little different, which is  $0.93 \text{ nm}^{-1}$  in the atomistic simulation and  $0.89 \text{ nm}^{-1}$  in the CG simulation with torsion potentials. Although this could cause deviation to some extent in  $S(q)$  plots, the positions of two main peaks in low and large  $q$  regimes obtained from CG simulation with torsion potentials are matched well to the ones from atomistic simulations, demonstrating that the CG lamella has quite similar structure as seen in the atomistic lamellae. The higher peaks obtained from the CG model with torsions can be explained by its overestimate of the bulk density at 300 K by a factor of 2% when compared to the atomistic target, which promotes a higher order degree of structure than is seen in the atomistic simulation.

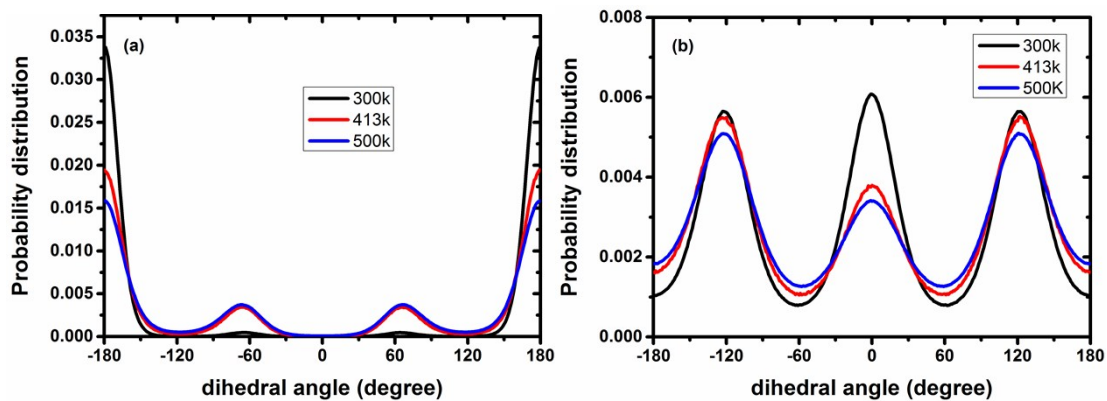


Fig. S4. Probability distribution function of dihedral angles of (a)  $\psi_1$  between united atoms CH-CH<sub>2</sub>-CH<sub>2</sub>-CH and (b)  $\psi_2$  between united atoms CH=CH-CH<sub>2</sub>-CH<sub>2</sub> at various temperatures in atomistic simulation.