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Monte Carlo simulations of star-branched polymers confined between two walls

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The properties of simplified lattice models of confined branched polymers were studied by the Monte Carlo method. Model chains were located between two parallel impenetrable surfaces. The distance between surfaces, as well as the chain length, were varied. The model chains consisted of $f=3$ branches of equal length (star-branched polymers) and were based on a simple cubic lattice. The model was athermal and the excluded volume was introduced. Monte Carlo sampling algorithm using local chain micromodifications was applied. Both static and dynamic properties of the system were investigated. The differences between star-branched chains and linear ones which were trapped between walls were discussed. The behavior of static properties made it possible to construct a universal curve presenting the effect of surface-to-surface distance on chain dimensions. The possible explanation of the shape of this curve was given. The changes of dynamic properties were discussed in the context of possible chain's mechanism of motion. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1429657]

INTRODUCTION

Polymers confined between two walls which were separated apart at the distance comparable with chain dimensions have recently been a subject of theoretical and experimental interest because of their practical significance (coating, laminates, lubrication, liquid crystals, biological systems).[1–3] The influence of the confinement on the size and shape of polymer systems has been recently presented in many theoretical works. Erman and Monnerie[4] and Cifra and Bleha[5] studied force-deformation relations in cases of a single chain as well as many-chain systems. The simulation of the block copolymer in the same confinement done by the means of computer simulation and using bond fluctuation model[6] showed that lamellae interfaces were perpendicular to the walls (neutral walls create ''entropic surface field''). Last year an interesting work was published[7] in which the influence of the presence of hard walls on the chain entanglements was studied for an off-lattice model of a polymer chain with the variable excluded volume.

Milchev and Binder[8] studied a single polymer off-lattice chain using the Monte Carlo method, they determined determining its structure and scaling relations. Shih *et al.*[9] studied the influence of the polymer concentration on the properties of systems near a hard wall. The results of these simulations were close to the scaling predictions of De Gennes.[10] Simulations of the confined dense polymer melt were carried out by Yoon *et al.*[11] and ten Brinke *et al.*[12] They showed that the static properties of a chain were disturbed over a length scale that was comparable to the doubled unperturbed radii of gyration. On the other hand, properties of many-chain systems were disturbed over much lower scale (comparable with three bond lengths). Very dense systems of linear chains were simulated by Pakula and Zhulina[13–15] who used a cooperative motion method. They found that hard walls had the significant influence on the chains' orientation but, at the same time, their influence on shapes was very small. The influence of the local stiffness of chains on the properties of melts was studied by Yethiraj.[16] Another recent work on chains between walls[17] contained the results obtained using a variational theory for hard (impenetrable) and soft (short-range repulsive) walls. They showed that the crossover from the three-dimensional to the two-dimensional state of a polymer chain was not monotonic while considering the size of the chain. The same behavior was observed in computer simulations.[18,19] Dickman *et al.* have performed Monte Carlo off-lattice and lattice simulations of oligomers in confined space. They studied the properties of the system in different solvent conditions. The derived the equation of state as well as the scaling exponents of the system.[20–23]

Dynamic properties of linear chains between hard walls were also studied recently. The review of the dynamics of polymer melts in slit has been given by Semenov *et al.*[24] Milchev and Binder[25] carried out Monte Carlo simulations of a polymer chain between impenetrable walls using an off-lattice model of a polymer chain. They found that, in general, a chain behavior was Rouse-type, i.e. the diffusion for short displacements was anomalous (diffusion coefficient scales with time as $D\sim t^{0.6}$) and in the free diffusion limit $D\sim N^{-1}$. It was surprising that the influence of the distance between walls on the diffusion coefficient was rather weak.

Some interesting questions concerning the properties of polymer chains trapped between two surfaces still remain unanswered. We know that in the case of free chains the presence of branching changes the structure of a polymer chain and thus its size, shape and viscoelastic properties.[26,27] It would be interesting to check if similar effects occur for a

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polymer trapped in a slit. The second interesting question concerns the influence of the confinement on the dynamic properties. The careful analysis of the changes in the dynamic behavior should give some insight into the mechanism of chain motion. Some initial results concerning the motion of star-branched polymers between impenetrable walls with obstacles were recently published.[28] It was shown that dynamic properties of such chains resemble those of the polymer melt.

In the present paper, we studied a simple lattice model of isolated star-branched polymers (infinitely diluted solution) confined between two hard walls by the means of a Monte Carlo method. The star-branched polymers were chosen as a subject of the study because they were not only idealized models useful for theoretical consideration but also could be synthesized and studied in real experiments.[26,27] The algorithm which was employed in the present study was developed specially for star-branched polymers[29,30] and used previously for free chains, melts, and adsorbed chains.[31–35] It is efficient for isolated chains even in the presence of an impenetrable surface as well as in dense systems.

The organization of this paper is as follows. In the next section we are describing the model and briefly the sampling algorithm. In the section ''Results and Discussion'' we are presenting the results concerning the size of chains and their scaling. Self-diffusion and relaxation of star polymers are also discussed in this section.

DESCRIPTION OF THE MODEL

In our simulations we have employed a star polymer molecule with $f=3$ arms of equal length n emanating from a common origin called a branching point. The total number of beads in the molecule was $N=f(n-1)+1$. The chains were constructed on a simple cubic lattice with the excluded volume effect present. The system was athermal, i.e., no long-range attractive and local potentials were present. Hence, our model simulates a good solvent conditions.[29,36]

We used the classical Metropolis scheme in order to sample efficiently the configurational space and to calculate some parameters describing the polymer size, shape, and dynamic properties. The initial configuration of a model chain underwent a series of local micromodifications where positions of a few beads (segments) were changed at random. It was proved that a proper set of such modifications allows a chain to get to any point in the phase space. We used a set of motions which was successfully used previously for linear chains: (i) Two-bond chain end motion, (ii) two-bond kink motion, (iii) three-bond kink motion, (iv) three-bond 90° crankshaft motion, (v) branching point collective motion. Collective means that the move of the branching point is accompanied by a move of an adjacent bead belonging to one of three arms. A detailed description of the above elementary motions was given elsewhere.[29]

The simulations were done for a series of different distances d (in lattice units) between the two impenetrable (pure repulsive) walls. The walls were parallel to the xy-plane and the z axis was perpendicular to the walls. The periodic boundary conditions were applied in x and y directions only. The scheme of the model was shown in Fig. 1. The smallest

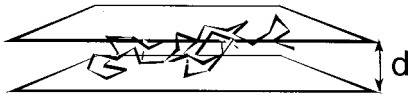

FIG. 1. The scheme showing the model of a star-branched polymer chain between two parallel impenetrable surfaces.

value of d in our simulations was three lattice units, i.e., the value at which all of the possible chain's micromodifications could take place.[29] Then, the distance d was varied in order to check the behavior of the system at larger d up to $d \gg \langle S^2 \rangle$. One can see that the limiting case $d=\infty$ equals to a chain without any confinements what corresponded to a free chain.

In order to confine the model macromolecules between two parallel impenetrable walls we started the first series of simulations at $d=3$ from a flat two-dimensional structure and then an equilibration run was performed. After the equilibration was done we performed a long run during which the trajectory was recorded. The typical trajectory consisted of 3000–5000 snapshots taken with the time step dependent on both: The chain length and d. Since the main goal of the simulations was to determine the static and dynamic properties of the system we had to sample the configurational space as well as to record the snapshots with the proper time step. The too large time step could cause the overlooking of the short time dynamics. On the other hand, too short time step caused that the self-diffusion process did not reach the region of the free diffusion. The simulations were repeated many times starting from different conformations. After the simulation run at certain d the output chain served as the input chain for new series of simulation at larger d. Each series of simulations was proceeded by the equilibration of the system.

RESULTS AND DISCUSSION

Static properties

We performed the series of simulations for star-branched polymers with the arm length $n=17$, 34, 67, and 134 what corresponded to total number of beads $N=49$, 100, 199 and 400, respectively. The simulations were carried out at different distance between the walls d starting from $d=3$ and then increasing this parameter up to $d=50$. The size of the star-branched polymer chain was described by the mean-square center-to-end distance $\langle R^2 \rangle$ and the mean-square radius of gyration $\langle S^2 \rangle$. We limited our considerations to the studies of influence of the distance between the walls on the properties of the chains since the detailed analysis of chain's shape and scaling behavior was recently published.[8,37]

The values of $\langle R^2 \rangle$ and $\langle S^2 \rangle$ parameters for various chain lengths and for different distances between the walls were listed in Table I. The standard errors of these quantities are less than 0.4% and 0.2%, respectively. Figure 2 showed the dependence of the mean-square center-to-end distance $\langle R^2 \rangle$ distance d on the separation d, for various chain lengths. It can be noticed, that for the small distance between the walls, the dimensions were relatively large, then, along with the increase of d they decreased gradually, reaching a

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TABLE I. Parameters describing the size of star-branched polymer chains.

	$N=49$		$N=100$		$N=199$		$N=400$	
D	$\langle R^2\rangle$	$\langle S^2\rangle$	$\langle R^2\rangle$	$\langle S^2\rangle$	$\langle R^2\rangle$	$\langle S^2\rangle$	$\langle R^2\rangle$	$\langle S^2\rangle$
3	33.3	14.0	91.3	38.4	250.9	104.0	708.7	290.8
5	29.9	12.4	75.1	31.6	199.0	82.7	534.3	217.3
7	29.7	12.3	72.0	28.6	176.2	73.1		
10	30.5	12.5	69.6	28.2	163.5	67.7	392.0	165.6
12	30.8	12.7	71.0	28.9				
15	31.6	13.0	72.8	29.6	159.1	64.2	383.5	153.0
20					160.8	64.9	371.2	147.0
30					169.8	68.5		

minimum value at certain d. The same behavior was observed in Fig. 3 which showed the dependence of the mean-square radius of gyration $\langle S^2\rangle$ as a function of the distance d. It was the effect which was observed very recently for the linear chains.[5,11,12,37] At small d, the chain was squeezed by the walls and, therefore, the dimensions were relatively large. Then, during the increase of the distance between the walls, the chain took the form of the coil what resulted in the decrease of the dimension parameters. It is obvious that the radius of gyration obeys the relation $\langle S^2\rangle=\langle S_x^2\rangle+\langle S_y^2\rangle+\langle S_z^2\rangle$—where the subscripts denoted the components calculated along the x, y, and z axis respectively. Therefore, one can write $\langle S^2\rangle=\langle S_\parallel^2\rangle+\langle S_\perp^2\rangle$, where subscript $_\parallel$ denotes the radius of gyration calculated with respect to xy-plane (parallel to the walls), while the subscript $_\perp$ denotes the component perpendicular to the walls (taken along the z axis). The same equation held the mean-square center-to-end distance $\langle R^2\rangle$. According to the above relation, in Fig. 4 we plotted the values of $\langle S_\parallel^2\rangle$ and $\langle S_\perp^2\rangle$ as a function of d. As one can see, the both terms behaved in an opposite way: The $\langle S_\parallel^2\rangle$ decreased while $\langle S_\perp^2\rangle$ increased. Since $\langle S^2\rangle$ was the sum of these two terms (one increasing, another decreasing along with the increase of d), it was not surprising that it exhibited the minimum. Finally, at very large d, the dimensions reached the parameters of the free chain. However, the $\langle S_\parallel^2\rangle$ did reach the free chain limit for smaller d than does $\langle S_\perp^2\rangle$. This could be explained as follows: There were no constrains in the xy plane of the model; the z direction was confined by the walls even for large d, therefore, this case could not be regarded as the "pure" free chain limit.

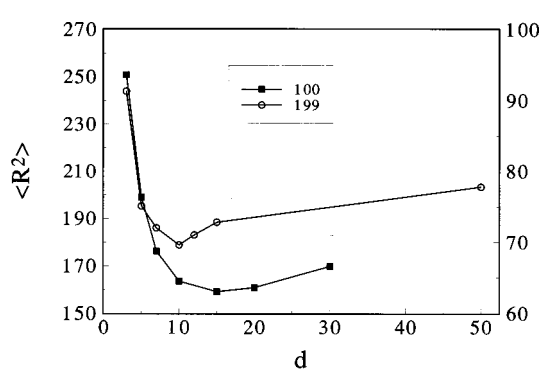

FIG. 2. The mean-square center-to-end distance $\langle R^2\rangle$ vs the distance between the walls d. The case $N=100$ and 199.

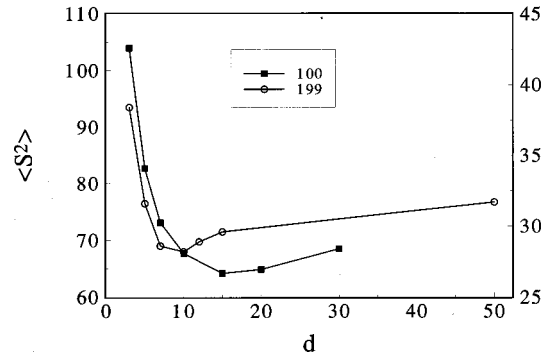

FIG. 3. The mean-square radius of gyration $\langle S^2\rangle$ vs the distance between the walls d. The case of $N=100$ and 199.

It was previously shown that this behavior was characteristic for a linear chain trapped between repulsive and soft walls although the worsening of the solvent led to the monotonical changes of chain's dimensions.[17] The location of the minima on $\langle S^2\rangle$ curves corresponded to the value of twice radius of gyration what was previously found in simulation for models of the melt of linear chains and of single linear chains.[5,11,12,37] The changes of the parallel $\langle S_\parallel^2\rangle$ and perpendicular $\langle S_\perp^2\rangle$ components of the mean-square radius of gyration were similar to those found by Cifra and Bleha for linear chains.[5] The component parallel to the walls $\langle S_\parallel^2\rangle$ increased almost twice as the effect of squeezing of the polymer takes place.

As one could expect, the position of the minima on the plots $\langle S^2\rangle$ and $\langle R^2\rangle$ vs d was related to both: The total length of the chain and the distance between the walls. If the distance d was smaller than the diameter of the coil, the chain would be squeezed by the walls and therefore its size became larger in the xy-plane. On the other hand, the z axis dimension was limited by d. The free chain coil had its dimension which we denoted as $\langle S^2\rangle_0$; this meant that the mean radius of the coil (treated as a sphere) was $\langle S^2\rangle_0^{1/2}$. Therefore, if d was smaller than approximately the doubled mean radius of the coil, the molecule was squeezed. At larger d the effect of squeezing vanished gradually, reaching values appropriate for a free chain. The effect of the changes in dimensions along with variation of d could be shown with respect to the unperturbed values of $\langle S^2\rangle_0$ as the ratio $\langle S^2\rangle/\langle S^2\rangle_0$. There-

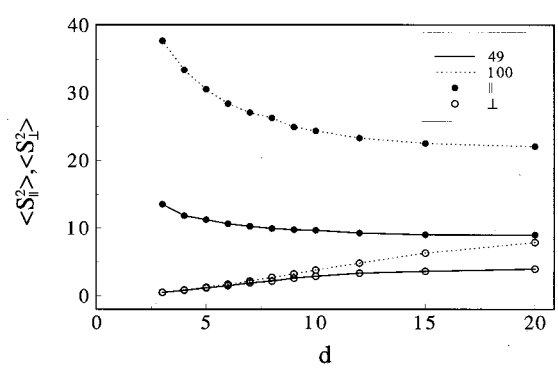

FIG. 4. The parallel $\langle S_\parallel^2\rangle$ and perpendicular $\langle S_\perp^2\rangle$ to the surface components of the mean-square radius of gyration vs the distance between the walls d. The case of $N=49$ and $N=100$.

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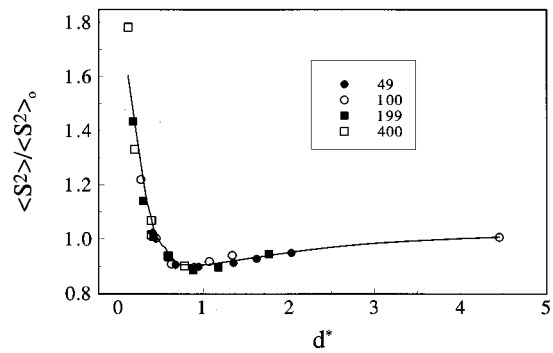

FIG. 5. The reduced mean-square radius of gyration $\langle S^2\rangle/\langle S^2\rangle_o$ vs reduced distance between walls d^* for the chains with $N=49$, 100, 199, and 400 beads.

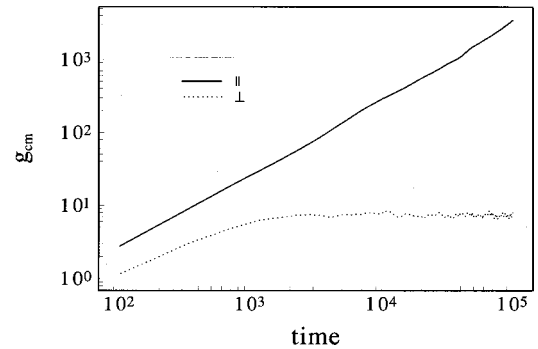

FIG. 7. The center-of-mass autocorrelation function g_{cm} split into parallel and perpendicular components for chains with $N=100$ and $d=15$.

fore, in Fig. 5, we plotted the latter quantity as a function of the dimensionless variable $d^*=d/(2^*(\langle S^2\rangle_0)^{1/2}$ for different chain length. The values of $\langle S^2\rangle_0$ were taken from Table I in Ref. 29. The obtained set of points formed a uniform curve with the characteristic minimum near value $d^*=1$, i.e., twice the radius of gyration of the free chain. The scaling distance similar to d^* was introduced by ten Brinke *et al.*[18] for linear chains. They used the distance parameter based on the polymer chain's size scaling exponent.

Investigating the model one could expect some similarity between the chain squeezed between the walls and the chain adsorbed on the planar surface. Recent Monte Carlo simulations showed that even strongly adsorbed star-branched chains exhibited the dimensions along z axis greater than the chains located between the two repulsive walls.[35] This showed that these two models described two different cases of confinement.

Dynamic properties

The short-time dynamics characterized the relaxation processes within the chain. We have investigated the autocorrelation function of the center-to-end vector. This procedure was described in detail elsewhere.[29,30] Then, from the center-to-end vector autocorrelation function g_R, the longest relaxation times τ_R were calculated according to the relation $g_R\sim\exp(-t/\tau_R)$. The values of τ_R were calculated with the standard error not greater than 4%. The plot of τ_R as a function of the distance between surfaces (Fig. 6) showed that the

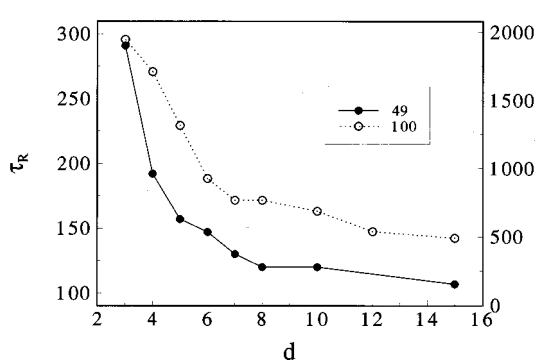

FIG. 6. The longest relaxation times τ_R versus the distance between walls d. The case of $N=49$ and 100.

relaxation processes were strongly affected by the confinement between the walls. At distances between the walls less than $2\langle S^2\rangle^{1/2}$ the relaxation time decreased sharply with the increase of d. The further increase of d caused the slow decrease of relaxation times to a final plateaulike region. The crossover between the two regimes was located close to the region of the reduced distance $d^*=1$. In this region, it could be observed that the curves were not smooth, as expected, because they lost their hyperbolic shape. The behavior of the dynamics in this region required further studies.

In order to characterize the long-time dynamics of the system we performed long simulations during which the trajectories of the system were recorded. The length of the trajectory depended on both—the length of the chain and the distance between the walls d. The trajectory had to be long enough to obtain the correct statistics of the translation of the chain. This was required in order to obtain the correct values of the diffusion constant. The procedure of extracting the diffusion coefficient was described in details elsewhere.[29,30] Here, we are presenting the main idea of it.

In order to investigate the influence of the confinement on the chain's diffusion we discuss the two-dimensional diffusion (in xy direction) only. This is because the motion along the z axis was limited by the presence of impenetrable surfaces. The center-of-mass (cm) autocorrelation function defined as $g_{\mathrm{cm}}(\Delta t)=<\mathbf{r}_{\mathrm{cm}}(t)\cdot\mathbf{r}_{\mathrm{cm}}(t+\Delta t)$, where $\mathbf{r}_{\mathrm{cm}}$ was a center-of-mass vector and t denoted time. Figure 7 presents an example of the function g_{cm} split into xy and z contributions. It can be seen that the z component of g_{cm} reached a plateau at the distance $d/2$. The behavior of xy-component suggested the Rouse-type motion what was previously found for linear chains.[32] The two-dimensional diffusion coefficient $D_{\parallel}$ was defined according to Einstein's relation as $D_{\parallel}=g_{\mathrm{cm}\parallel}(t)/4t$ for sufficiently long times,[29,30] where $g_{\mathrm{cm}\parallel}\sim t^1$. The values of D were calculated with the standard error not greater than 2%.

In order to determine the influence of d on diffusion of the system we have plotted the diffusion coefficient ratio $D_{\parallel}/D_{\parallel}^o$ where $D_{\parallel}^o$ denoted the diffusion coefficient of the free chain (but calculated for two dimensions only) against d^* for different chain lengths (the values of $D_{\parallel}^o$ were taken from the Table II of Ref. 29). It can be seen that the obtained curves for different chain lengths were of similar shape. At values of d^* between 0 and 1 curves showed maximum value, then the

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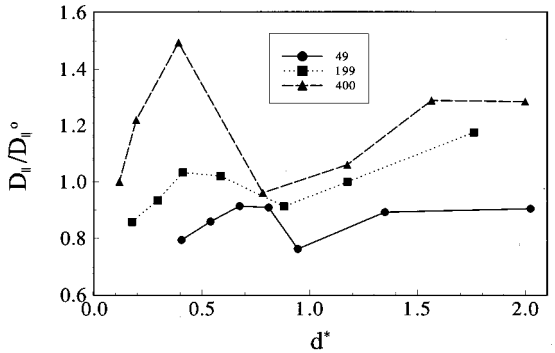

FIG. 8. Two-dimensional reduced self-diffusion coefficient $D_{\parallel}/D_{\parallel}^{o}$ vs the reduced distance between walls d^* for chains with $N=49$, 199, and 400 beads.

minimum reached in the vicinity of $d^*=1$ and next the curves increased when $d^*>1$. The position of the minima on curves depended on the chain length. The longer the chain the smaller were the values of d^* at which the position of the minimum was located. The minima on curves meant, that the diffusion in the region of d^* in the vicinity of 1 was slowed down. The resulting shape of the curves was the result of a complicated phenomenon, which was governed by many factors, like: The confinements of the chains, the excluded volume effect, the segment-density distribution and the mechanism of motion. The analysis of the curves presented in Fig. 8 showed that the diffusion coefficient at $d^*>1$ increased along with the increase of the distance between the walls. In this region larger d^* increased effectiveness of the micromodifications of the chain and, therefore, enabled the greater mobility of the system. However, the further increase of the parameter d^* did not lead to the significant enhancement of the diffusion.

From Fig. 8 it can be seen that diffusion coefficient gradually increased as d^* became less than 1. In our opinion at least two factors caused this effect: The change in the mechanism of the chain motion and probably less effective disentanglement processes. As it could be expected, the flattened star-branched chain strongly reduced the number of effective micromodifications and probably the mechanism of motion changed as the system obtained more freedom at larger d^*. On the other hand, as the chain became less squeezed between the two walls, the mobility of the chain branches increased. The mechanism of motion was strongly connected with the entanglements of the chain.[36] Here, we expected, that the number of entanglements in the chain which was strongly squeezed between the walls $(d^*<1)$ was reduced while compared it to the conditions for $d^*>1$. On the other hand, the *existing* entanglements were more stable in the flattened chain confined between the impenetrable walls. Further increase of d^* from 1 up to 2 led to the increase of the ratio of diffusion coefficients $D_{\parallel}/D_{\parallel}^{o}$. It is interesting that this ratio was well over unity in this region what implied that these chains were more mobile then the unconfined ones. This surprising phenomenon could be explained by the fact that the chains under consideration apparently contained fewer entanglements than the free chains. The was probably caused by the fact that, as it was described above, we started the simulations from flat two-dimensional (2D) structures, which had no entanglements. The increase of d did not lead to the normal number of chain entanglements because of existing space limitations (the presence of walls). This explanation could be confirmed by the fact that shortest chains, where the number of entanglements was much lower, achieved the free chain diffusion limit ($D_{\parallel}/D_{\parallel}^{o}$ close to unity). The above discussion suggests that detailed studies of confined chain dynamics including the opposite course in changing the distance between walls could give some insight into the nature of chain entanglements. The appropriate studies are underway.

The above-discussed static and dynamic properties of star-branched polymer chains confined between two parallel impenetrable surfaces appeared to be a very interesting subject of theoretical studies. It was shown that hard walls significantly influence the static and dynamic properties of a polymer chain. It was interesting that these dependencies could be described by one universal curve. The proper scaling of parameters describing the model system helped to predict a behavior of the chain at given geometrical constraints.

The changes of chain's size and shape with the distance between surfaces were connected with the transition from three-dimensional to two-dimensional system. Dynamics of the polymer chain was found very sensitive to this constraint too, although this dependence has a nonmonotonical character. This could be probably explained by the changes in the chain's mechanism of motion.

[1] *Physics of Polymer Surfaces and Interfaces*, edited by I. C. Sanchez (Butterworth-Heinemann, Boston, 1992).
[2] G. J. Fleer, M. A. Cohen-Stuart, J. M. H. M. Scheutjens, T. Cosgrove, and B. Vincent, *Polymers at Interfaces* (Chapman and Hall, London, 1993).
[3] R. Eisenriegler, *Polymers near Surfaces* (World Scientific, Singapore, 1993).
[4] B. Erman and L. Monnerie, Macromolecules **30**, 5075 (1997).
[5] P. Cifra and T. Bleha, Macromol. Theory Simul. **8**, 603 (1999).
[6] J.-U. Sommer, A. Hoffmann, and A. Blumen, J. Chem. Phys. **111**, 3728 (1999).
[7] G. Arteca, Macromol. Theory Simul. **8**, 137 (1999).
[8] A. Milchev and K. Binder, Eur. Phys. J. B **3**, 477 (1998).
[9] W. Y. Shih, W.-H. Shih, and I. A. Aksay, Macromolecules **23**, 3291 (1990).
[10] P.-G. de Gennes, *Scaling Concepts in Polymer Physics* (Cornell University Press, Ithaca, NY, 1979).
[11] S. K. Kumar, M. Vacatello, and D. Y. Yoon, J. Chem. Phys. **89**, 5206 (1988).
[12] G. ten Brinke, D. Ausserre, and G. Hadziioannou, J. Chem. Phys. **89**, 4374 (1988).
[13] T. Pakula, J. Chem. Phys. **95**, 4685 (1991).
[14] T. Pakula and E. B. Zhulina, J. Chem. Phys. **95**, 4691 (1991).
[15] E. B. Zhulina and T. Pakula, Macromolecules **25**, 754 (1992).
[16] A. Yethiraj, J. Chem. Phys. **101**, 2489 (1994).
[17] C. E. Cordeiro, M. Molisana, and D. Thirumalai, J. Phys. II **7**, 433 (1997).
[18] J. H. van Vliet and G. ten Brinke, J. Chem. Phys. **93**, 1436 (1990).
[19] J. H. van Vilet, M. C. Luyken, and G. ten Brinke, Macromolecules **25**, 3802 (1992).
[20] R. Dickman and C. K. Hall, J. Chem. Phys. **89**, 3168 (1988).
[21] A. Hertanto and R. Dickman, J. Chem. Phys. **89**, 7577 (1988).
[22] R. Dickman, J. Chem. Phys. **91**, 454 (1989).
[23] R. Dickman, J. Chem. Phys. **96**, 1516 (1992).
[24] A. N. Semenov, J. F. Joanny, A. Johner, and J. Bonet-Avalos, Macromolecules **30**, 1479 (1997).
[25] A. Milchev and K. Binder, J. Phys. II (France) **6**, 21 (1996).
[26] G. S. Grest, L. J. Fetters, J. S. Huang, and D. Richter, Adv. Chem. Phys. **94**, 67 (1996).

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[27] J. J. Freire, Adv. Polym. Sci. **143**, 35 (1999).
[28] P. Romiszowski and A. Sikorski, Macromol. Symp. **171**, 63 (2001).
[29] A. Sikorski, Makromol. Chem., Theory Simul. **2**, 309 (1993).
[30] P. Romiszowski and A. Sikorski, J. Chem. Phys. **104**, 8703 (1996).
[31] A. Sikorski, A. Kolinski, and J. Skolnick, Macromol. Theory Simul. **3**, 715 (1994).
[32] A. Sikorski and P. Romiszowski, J. Chem. Phys. **109**, 2912 (1998).
[33] P. Romiszowski and A. Sikorski, J. Chem. Phys. **109**, 6169 (1998).
[34] A. Sikorski and P. Romiszowski, Macromol. Theory Simul. **8**, 109 (1999).
[35] A. Sikorski, Macromol. Theory Simul. **10**, 38 (2001).
[36] J. Skolnick and A. Kolinski, Adv. Chem. Phys. **78**, 223 (1990).
[37] A. E. Giesen and I. Szleifer, J. Chem. Phys. **102**, 9069 (1995).

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