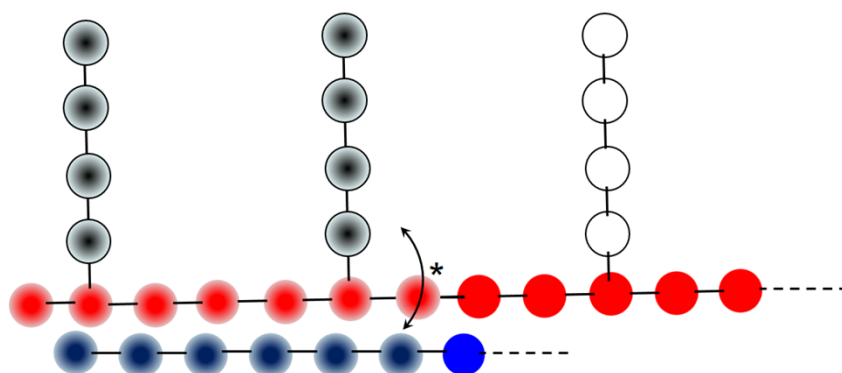


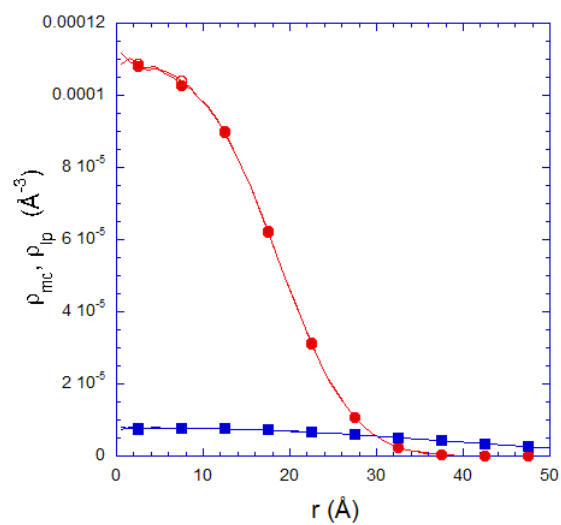
## ESI – Electronic Supplementary Information

### Branched–linear polyion complexes investigated by Monte Carlo simulations

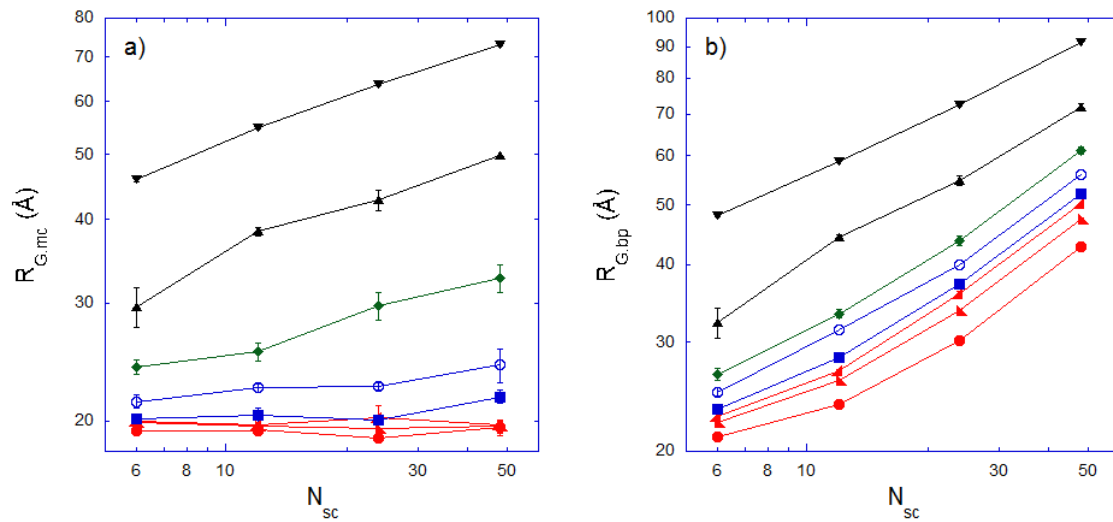
Daniel G. Angelescu and Per Linse



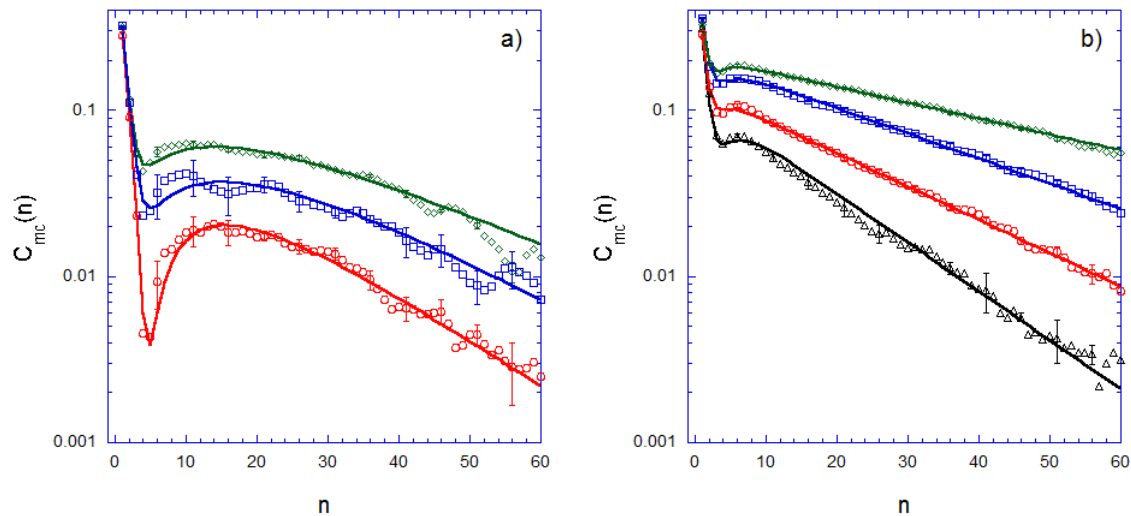
**Fig. S1** Schematic illustration of a trial pivot rotation. The pivot bead of the main chain was randomly chosen and the collective rotation comprises (i) a part of the main chain and (ii) side chains of the branched polyion attached to (i) and (iii) a part of the linear polyion located near the branched polyion. In the last case, the bead closest to the pivot bead divided the linear polyion in two parts, and the part near (i) was considered. The displacement parameter was a maximal angle of rotation, which was between  $30^\circ$  and  $90^\circ$ , and the rotation axis was given by the bond joining the pivot bead and its non-moving neighbor. The pivot bead is marked with \*, and the beads to be rotated are given in gradient solid. Same color code as in Fig. 1 is used.



**Fig. S2** Bead number density of the main chain of the branched polyion (solid symbols) and of the linear polyion (open symbols) as a function of the distance to the center of the mass of the branched polyion at the side-chain length  $N_{sc} = 12$  and the branching densities  $\sigma_b = 0.0208$  (●) and  $0.50$  (■). In nearly all cases, the open symbols are completely hidden by the solid symbols.



**Fig. S3** Rms radius of the gyration of (a) main chain  $R_{G,mc}$ , and (b) branched polyion  $R_{G,bp}$  as function of side chain length at the branching density  $\sigma_b = 0.0208$  ( $\bullet$ ), 0.0416 ( $\boxtimes$ ), 0.0625 ( $\boxplus$ ), 0.0833 ( $\blacksquare$ ), 0.125 ( $\circ$ ), 0.166 ( $\star$ ), 0.25 ( $\blacksquare$ ), and 0.50 ( $\square$ ).



**Fig. S4.** Bond-angle correlation function of the main chain of the branched polyion  $C_{mc}(n)$  (symbols) and the least-square fits to tri-exponential function (dotted curves) at the branching densities (a)  $\sigma_b = 0.25$  and (b)  $\sigma_b = 0.50$  and the side-chain length  $N_{sc} = 6$  ( $\square$ ), 12 ( $\circ$ ), 24 ( $\triangle$ ), and 48 ( $\ast$ ). Data for  $\sigma_b = 0.25$  and  $N_{sc} = 6$  are excluded, since the correlation function was continuously decaying.