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Interactions between Rigid Polyelectrolytes Mediated by Ordering and Orientation of Multivalent Nonspherical Ions in Salt Solutions

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Multivalent ions in solutions with polyelectrolytes (PEs) induce electrostatic correlations that can drastically change ion distributions around the PEs and their mutual interactions. Using coarse-grained molecular dynamics simulations, we show how in addition to valency, ion shape and concentration can be harnessed as tools to control rigid like-charged PE-PE interactions. We demonstrate a correlation between the orientational ordering of aspherical ions and how they mediate the effective PE-PE attraction induced by multivalency. The interaction type, strength, and range can thus be externally controlled in ionic solutions. Our results can be used as generic guidelines to tune the self-assembly of like-charged polyelectrolytes by variation of the characteristics of the ions.

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Electrostatic interactions between charged molecules and ions in solution are ubiquitous in colloidal, soft, and biological systems [1]. Systems such as some polyelectrolytes (PEs), synthetic and biopolymers, DNA [2,3], nanotubes in phospholipids [4], actin filaments [5,6], microtubules [7], viruses [8,9], and even bacteria [10], can often be approximated by charged cylinders immersed in an electrolyte solution consisting of a solvent and mobile ions [11]. Understanding the ion distribution in such systems is paramount since solution-mediated interactions are greatly affected by the ionic environment, especially due to electrostatic screening effects [12] and ion redistribution [13,14]. Both the nature and concentration of ions play a significant role, whence ion valency is an important handle for tuning the properties of macroions [15–19].

Many chemically specific ions, such as, e.g., diamine, spermine, and spermidine, exhibit elongated, cylindrical shapes and are multivalent [7,20]. Some anions in battery electrolytes are nonspherical, which influences ion transport and conductivity in solution [21,22]. Ionic liquids are typically composed of highly nonspherical ions, which

influences their cohesion energy and maintains their liquid character, but also influences ionic transport [22,23]. Consequently, ion specificity is paramount in controlling interactions between charged macromolecules.

For modeling purposes, traditional mean-field approaches such as Poisson-Boltzmann (PB) theory treat mobile ions as point charges in the weak-coupling regime. The standard PB theory and extensions to it [24] cannot describe general chemically specific ions [25,26]. However, successful models incorporating ion size properties exist, such as those in Refs. [27–32]. In the case of like-charged PE-PE interactions, the PB theory always predicts repulsion. To this end, the soft-potential-enhanced-PB (SPB) theory has been shown to accurately predict ion distributions around PEs [33] for ion sizes up to the PE radius, and like-charged PE-PE repulsion for small monovalent ions such as Na⁺ and Cl⁻ [34]. In Ref. [35], however, like-charged PE-PE attraction was reported for large monovalent ions and high salt concentration, and attributed to short-range charge correlations beyond the PB theory.

For multivalent ions, charge-charge correlations naturally appear and cause charge reversal (see, e.g., Refs. [36– 38]) in the strong-coupling regime even with pointlike ions [15,39–47]. Both valency and ion size have been considered in the context of classical density functional theory [48]; see, e.g., the recent advances on electric double layers

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[49–51]. There also exist Monte Carlo studies on dumbbelllike (two separated point charges), yet volumeless, ions focused on counterion-mediated interactions between charged plates [52–57] or cylinders [58]. References [56,58–63] have suggested that a bridging mechanism is responsible for the attraction between like-charged surfaces. Nevertheless, to our knowledge none of these approaches have simultaneously considered both correlations and steric effects of aspherical multivalent ions.

In this Letter, we extend the ion-mediated interaction scenario in the case of spheroidal multivalent ions. Using coarse-grained (CG) molecular dynamics (MD) simulations, we focus on systems composed of single and double rodlike PEs. We first investigate the condensation and orientation response for different ion specificities around a single PE. We then address the order-mediated interactions between two like-charged PEs and underpin the effect of ion orientation, valency, and shape. Tuning the balance between attractive and repulsive forces allows for controlling the thermodynamic properties of assembled systems, their stability, and their response to external conditions [7,8,20,64–67]. We show how this control can be achieved by adjusting ion valency, shape, and salt.

Model and theory.—The setup consists of a periodic cubic box filled with charged spheroidal mobile ions and one (or two) fixed charged rod(s). The simulations are carried out in the *NVT* ensemble where we employ the Nosé-Hoover thermostat [68,69] with a coupling constant of 0.2 ps and reference temperature T = 300 K. The equations of motion are integrated using a velocity Verlet algorithm with a time step of 2 fs to satisfy energy conservation. The production run lasts 20 ns, out of which the first 5 ns are omitted in the data analysis (equilibration). The initial configurations are prepared using MOLTEMPLATE [70] and PACKMOL [71]. Figure 1 shows the schematic representation of our model.



FIG. 1. Ions with (a) $A^c = 1$, (b) $A^c = 2$, (c) $A^c = 3$, and (d) $A^c = 4$. The charges are separated from the center by $\sigma_{maj}/4$ (green dots). (e) Snapshot of the simulation box of size (20 nm)³ with periodic boundary conditions containing one PE (grey), cations (red), and anions (cyan), where $A^c = A^a = 3$.

The LAMMPS software was used for the simulations [72,73]. The interactions between components i and j (also between PE and ions) at a distance r are modeled via a soft repulsive version of the orientation-dependent Gay-Berne potential [74,75], which is obtained by shifting and truncating the potential as

$$U^{ij}(\hat{u}^{i}, \hat{u}^{j}, r^{ij}) = \epsilon^{ij}(\hat{u}^{i}, \hat{u}^{j}, \hat{r}^{ij})[4(\Sigma_{ij}^{12} - \Sigma_{ij}^{6}) + 1] \quad (1)$$

at $r^{ij} < r_c^{ij}(\hat{\boldsymbol{u}}^i, \hat{\boldsymbol{u}}^j, \hat{\boldsymbol{r}}^{ij})$, where

$$\Sigma_{ij} = \frac{\sigma_0^{ij}}{r^{ij} - \sigma^{ij}(\hat{\boldsymbol{u}}^i, \hat{\boldsymbol{u}}^j, \hat{\boldsymbol{r}}^{ij}) + \sigma_0^{ij}}.$$
 (2)

Here $\hat{\boldsymbol{u}}^i$ and $\hat{\boldsymbol{u}}^j$ are the unit vectors along the molecular axes, σ_0^{ij} the minimum contact distance for the ij pair, σ^{ij} the orientation-dependent separation distance at which attractive and repulsive contributions cancel, $\epsilon^{ij} = \epsilon_0^{ij} [\epsilon^{ij} (\hat{\boldsymbol{u}}^i, \hat{\boldsymbol{u}}^j)]^{\nu} [\tilde{\epsilon}^{ij} (\hat{\boldsymbol{u}}^i, \hat{\boldsymbol{u}}^j, \hat{r}^{ij})]^{\mu}$ the orientation-dependent well depth, and $r_c^{ij} (\hat{\boldsymbol{u}}^i, \hat{\boldsymbol{u}}^j, \hat{r}^{ij})$ the position of the potential minimum [see Supplemental Material (SM) [76]]. Following Ref. [77] we set $\nu = 1$ and $\mu = 2$.

We set $\sigma_0^i = 0.4$ nm (common hydrated diameter of ions) and $\epsilon_0^i = 0.1$ kcal mol⁻¹. The major (σ_{maj}) and minor (σ_{min}) axes define the aspect ratio $A = \sigma_{maj}/\sigma_{min}$. We set $\sigma_{maj} = \sigma_0^i A^{2/3}$ and $\sigma_{min} = \sigma_0^i A^{-1/3}$, which, regardless of *A*, provides a volume equivalent to the one of a sphere (A = 1) with diameter equal to σ_0^i . Here we consider A = 1, 2, 3, and 4. The choice of $\epsilon_{maj/min}^i$ follows Ref. [78].

The rigid PE is built of charged spherical beads (force centers) interacting via the Weeks-Chandlers-Andersen [79] potential $U^{\text{PE}}(r) = 4\epsilon_{\text{PE}}[(\sigma_{\text{PE}}/r)^{12} - (\sigma_{\text{PE}}/r)^6] + \epsilon_{\text{PE}}$ for $r \leq 2^{1/6}\sigma_{\text{PE}}$. Here, the bead diameter is $\sigma_{\text{PE}} = 1.2$ nm, and the depth of the potential well ϵ_{PE} is equal to ϵ_0^i . The beads are fixed at a distance b = 0.27 nm apart so that a smooth equipotential surface is experienced by the ions. The PE dimensions are in line with common synthetic and biopolymers, such as poly(styrene sulfonate) (PSS). Lorentz-Berthelot mixing rules $\epsilon_0^{ij} = \sqrt{\epsilon_0^i \epsilon_0^j}$ and $\sigma_0^{ij} = (\sigma_0^i + \sigma_0^j)/2$ are used. We use 74 beads, each with charge $Z^B e = -e$, providing a line charge density $\lambda = Z^B e/b \approx -4 e/\text{nm}$, close to that of PSS (-3.7 e/nm). The surface charge density $\lambda/\pi\sigma_{\text{PE}}$ is close to that of DNA molecules $(-1 e/\text{nm}^2)$.

The electrostatic interactions are modelled via Coulombic potentials, which, for two charges $Z^i e$ and $Z^j e$, read $\beta e V^{ij}(r) = Z^i Z^j \ell_B / r$, where $\beta = 1/k_B T$, and the Bjerrum length $\ell_B = \beta e^2 / (4\pi \varepsilon_r \varepsilon_0) = 0.7$ nm. In this, ε_r and ε_0 denote, respectively, the solvent dielectric constant (for water, $\varepsilon_r = 78$ at 300 K and 1 atm [80]) and the vacuum permittivity.

These contributions are obtained in reciprocal space, after a real space cutoff of 1.2 nm, using the particleparticle particle-mesh summation method [81] with relative force accuracy of 10^{-5} . Monovalent, divalent, and trivalent charges are equally split into two points at distances of $\sigma_{maj}/4$ from the center of the ions along the major axis, as sketched in Fig. 1. Finally, for valency (*Z*) and aspect ratio (*A*) of cations (c) and anions (a) we use the notation $Z^c: Z^a$ and (A^c, A^a) , respectively.

The PE is neutralized with counterions from multivalent added salt. The case of monovalent counterions with multivalent added salt is discussed in the SM. An extra monovalent anion is added for systems containing trivalent salt counterions. Finite size effects are checked by repeating the simulations for boxes with sides of 4, 6, 10, 20, 40, and 60 nm. All density profiles converge for boxes with sides of 20 nm.

The single-charge number density distribution of species i is obtained via

$$n^{i}(r) = \left\langle \sum_{k=1}^{N^{i}} \frac{Z^{i}}{2} \delta(|\boldsymbol{r} - \boldsymbol{r}_{k}^{i}|) \right\rangle_{t} \middle| V_{k}(r), \qquad (3)$$

where $\mathbf{r} = (x, y)$ is the distance vector on the *xy* plane from the center of the backbone of the PE, N_i the number of charges of type $i, \langle \cdots \rangle_t$ the time average, $\mathbf{r}_k^i = (x_k^i, y_k^i)$ are planar vectors pointing on single charges, and $V^k(r)$ the volume of a cylindrical shell located at *r*. Hereafter, cation and anion charge densities are denoted as n^+ and n^- , respectively.

To characterize the orientation along the PE z axis, we define the order parameter $S(r) \equiv 2 \langle |\hat{\boldsymbol{e}}_{z} \cdot \hat{\boldsymbol{u}}_{k}| \rangle_{t,r} - 1$, where $\langle \cdots \rangle_{t,r}$ is both time average and average over particles at r, \hat{u}_k the unit vector along the major axis of the kth ion, and \hat{e}_z the unit vector along the z axis. For ions oriented perpendicular to the PE S = -1, parallel S = 1, and randomly oriented S = 0. As an additional measure of the tendency of the ions to be tangential to the PE surface, we define $C(r) \equiv 1 - 2 \langle |\hat{\boldsymbol{e}}_r \cdot \hat{\boldsymbol{u}}_k| \rangle_{t,r}$, where $\hat{\boldsymbol{e}}_r$ is the unit vector normal to the PE surface. If all ions are tangent to the PE surface C = 1, if perpendicular C = -1, if randomly oriented C = 0. Finally, to quantify the tendency to orient along the x axis we use $\chi(\eta) = 2\langle |\sin \theta(\eta) \cos \varphi(\eta)| \rangle_{t,\eta} - 1$, where θ and φ are defined in Fig. 2(b) and $\eta = (x, y)$. Specifically, $\chi = 1$ for parallel, $\chi = -1$ for perpendicular, and $\chi = 0$ for random orientations. Figure 2 shows different ion orientations and the respective values of S, C, and χ .

Results for a single PE.—We first focus on the case of ion condensation around a single PE. We have recently shown that, for monovalent salt, a soft-potential-modified PB theory gives accurate results in the case of a cylindrical PE for a wide range of salt and ion sizes [33]. When multivalent ions are introduced into the system, such mean-field approximation breaks down. To this end, we have



FIG. 2. (a) Illustration of \hat{e}_r , \hat{u} . \hat{e}_r is normal to the cylinder surface, whereas \hat{u} points along the ion major axis. \hat{u} defines the orientation of the ion and characterizes the relative orientation with respect to the PE via $\hat{e}_r \cdot \hat{u}$. Configurations relative to the PE surface and their orientation order parameters *C* and *S*. (b) Order parameter χ is defined between two PEs. $\chi = 1$ if the ion is parallel to the *x* axis and $\chi = -1$ if perpendicular.

considered three different cases at ionic strengths of 0.5 M in detail. (i) Trivalent cations with spherical monovalent anions, i.e., case 3:1 and $A^a = 1$ with $A^c = 1-4$. (ii) Trivalent cations and anions, i.e., case 3:3 with $A^a = 3$ and $A^c = 2$, 3, 4. (iii) Trivalent cations and anions (3:3) with $A^c = 3$ and $A^a = 2$, 3, 4. Additional data for the effect of the ionic strength *I*, Z^c , and λ are shown in Figs. S2, S3, and S4 of SM, respectively.

The strong electrostatic attraction between the multivalent cations and the PE results in overcharging, as shown in Refs. [15,82,83] and a large peak in n^+ . The excess charge attracts anions, resulting in the formation of a second layer. This can be clearly seen in Figs. 3(a), 3(b), 4(a), and 4(b), where we show results for the cases (i) and (ii), respectively. Results for (iii) are very similar to (ii) and are shown in Fig. S6.

Interestingly, the n^+ data of the case (i) shows the aspherical cations have a much lower density near the PE surface than the spherical ones. This is in line with Monte Carlo simulations for dumbbell-like ions [52,56]. Furthermore, increasing the spacing between charges in



FIG. 3. (a) n^+ and (b) $|n^-|$ ($e \text{ nm}^{-3}$) in 3:1 salt with ionic strength of 0.5 M, $A^a = 1$ and varying A^c . (c) Order parameters $S^c(r)$ and (d) $C^c(r)$ for the systems shown in (a). The gray bar indicates the PE with a radius of 0.6 nm. Error bars in this and the other figures are comparable to the symbol sizes or smaller.



FIG. 4. (a) $|n^+|$ (solid symbols) and $|n^-|$ (open symbols) (e/nm^3) in 3:3 salt with ionic strength of 0.5 M, $A^a = 3$ and varying A^c . Panels (b)–(e) present the order parameters $S^a(r)$, $C^a(r)$, $S^c(r)$, and $C^c(r)$ for the systems in (a). The gray bar indicates the PE with a radius of 0.6 nm. (f) Schematic of cations with different anisotropy oriented perpendicular to the PE, showing that the negative minima are close to $\sigma_{\text{mai}}/2$.

the cations decreases the charge density close to PE (cf. Fig. S7).

The position of the spheroidal cation peaks indicates orientational ordering near the PE. To quantify this, the two order parameters are shown in Figs. 3(c), 3(d), 4(c), and 4(d). They indicate that the spheroids have a tendency to align along the backbone of the PE, as expected from electrostatics. Such ordering is enhanced with increasing electrostatic interactions, as shown in Fig. S4. Curiously, the order parameters show small negative minima indicating a tendency to align perpendicular to the PE. For case (ii), the closest spheroidal anions show a tendency to align with the cations while exhibiting no minimum. The positions of the order-parameter minima for cations correspond to $r \approx (\sigma_{\rm PE} + \sigma_{\rm maj})/2$, as shown in Fig. 4(f).

Results for PE-PE interactions.-We now turn to the interesting question of how multivalent spheroidal ions influence PE-PE interactions. In Ref. [34], we investigated the interactions between two negatively charged rods in monovalent spherical salt solutions, where the interactions are always repulsive for small ions when there are no correlations. Various strong-coupling approaches and Monte Carlo simulations have shown that charge correlations often lead to effective charge reversal of the PE and attractive interactions between them [35,58,62,84–86]. To quantify the PE-PE interactions, we have computed the mean force f(D) = F(D)/L (L being the PE length) and the corresponding potential $V(D) = \int_D^\infty F(x') dx'$ as a function of the surface-to-surface PE distance D. Here F(D) is the time-averaged total force acting between the PEs over production runs of 10 ns for discrete values of D. In Fig. 5 we show the results for spheroids with $A^c = 2-4$.



FIG. 5. Normalized mean force f (kcal mol⁻¹ nm⁻²) (first row), and βV (second row). The vertical solid lines indicate σ_{maj} and the dashed lines $(\sigma_{min} + \sigma_{maj})/2$ for $A^c = 2$ (light grey), $A^c = 3$ (grey) and $A^c = 4$ (dark grey), respectively. Error bars are comparable to symbol sizes.

As expected, the interactions for monovalent cations remain repulsive, and the cation shape and size have only a small influence [34].

The situation drastically changes when multivalent counterions are introduced. The forces for divalent and trivalent cations for different shapes are shown with green and cyan dots in Fig. 5. Here both charge correlations and ion shapes play systematically an important role. A deep negative minimum in the interaction potential appears, indicating strong binding. The values of the binding energy, together with the position of the minimum of f and of V, can be found in Table S1. The first notable result is that the minimum in f(D) approaches $D \approx \sigma_{maj}$ with increasing A^c and Z^c . Second, the equilibrium position corresponding to the potential minimum settles at $\approx (\sigma_{min} + \sigma_{maj})/2$. We note that even within the relatively limited range of aspect ratios studied here with $Z^c = 3$, the effect is significant as the attraction strength can be tuned by up to $\approx \pm 17\%$.

In Fig. 6, we show contour plots of the order parameter χ^c at values of D where the attractive force is largest. They reveal an interesting correlation to the cation orientation. Similar to the single-PE case, cations close to the PEs tend to align tangentially, whereas, in the region between the two PEs, parallel to the $x \, axis (\chi > 0)$. The middle and right columns of Fig. 6 show that the cations bridge the PEs at the minimum $D \approx \sigma_{maj}$, mediating attraction between the PEs [58,59,61,87,88]. The attractive region gradually moves toward higher values of D as A^c is increased since longer spheroids need more space to fit between the PEs. Larger values of Z^c lead to stronger electrostatic interactions, thus stronger attraction, but also to slightly smaller optimal distances between the PEs.

The corresponding contour plots at the equilibrium distance f = 0 are shown in Fig. S8. Interestingly, the orientational ordering of the cations between the PEs diminishes with increasing valency. This clearly demonstrates the role of orientation in mediating the forces. At the



FIG. 6. Contour plots of χ^c at the maximum attractive force (for $Z^c = 1$ the distance from $Z^c = 2$ is used). Contour plots at the equilibrium distance are shown in Fig. S8.

equilibrium distance, the cation-mediated attraction and the PE-PE repulsion exactly cancel out, and cations with higher valency require less ordering to neutralize the repulsion.

To study the influence of salt, we added monovalent 1:1 salt ($A^c = A^a = 3$) to the aforementioned systems, and the results are shown in Fig. S10. The attraction between PEs reduces because electrostatic interactions between them weaken. An interesting opposite case is that of monovalent counterions with multivalent salt in Fig. S11. The trivalent cations immediately lead to an attraction as charge correlations build up. The attraction is only weakly affected by added 3:1 salt up to 1 M.

Summary and conclusions.—In this Letter, we have shown how in addition to valency, both ion shape and concentration can be harnessed as tools to control likecharged PE-PE interactions. Multivalent ions induce an attractive force [58,62,65,84–86] whose magnitude and range can be tuned by the characteristics of the ions. In particular, we found a direct correlation between the orientational ordering of aspherical multivalent ions and the mediation of the attraction. Our results can be used as general guidelines to tune self-assembly by varying the ion properties.

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