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Supporting Information for

Monte Carlo Simulations of Ion Correlations and Transport in Highly Concentrated Liquid Electrolytes

Tabita Pothmann, Bernhard Roling

Department of Chemistry and
Marburg Center for Quantum Materials and Sustainable Technologies (mar.quest),
University of Marburg, Hans-Meerwein-Strasse 4, 35032 Marburg, Germany

Exemplary results for time-dependent transport coefficients

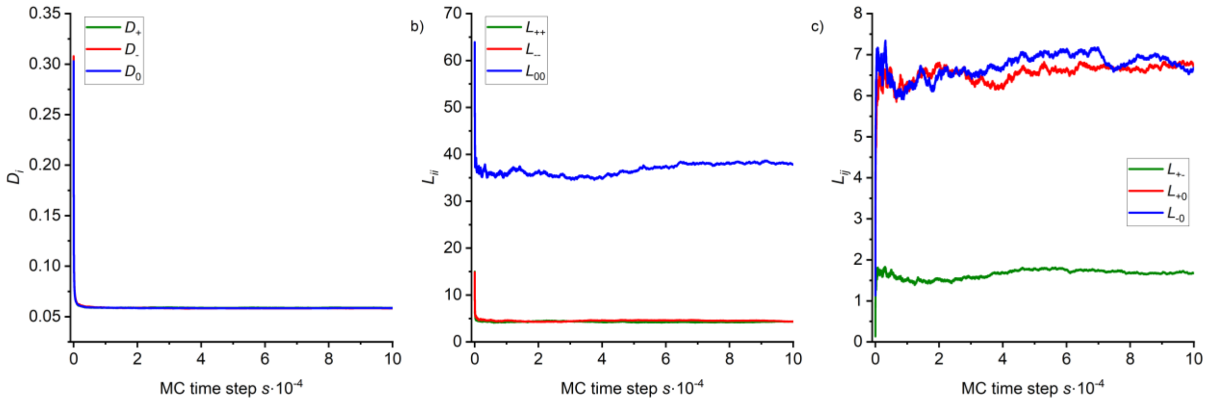

Fig. S1: Time-dependent diffusion coefficients and Onsager coefficients for a system with $1/x = 0.25$ and only hard-core interactions.

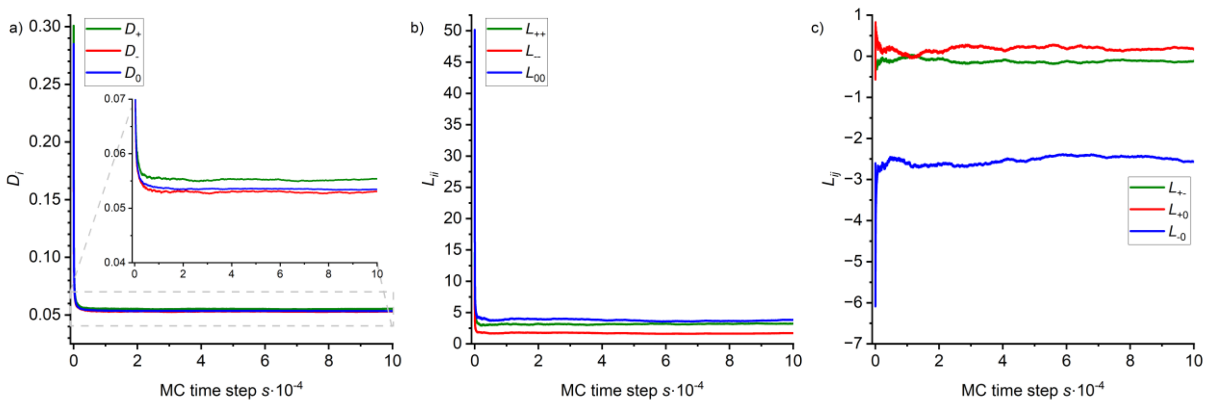

Fig. S2: Time-dependent diffusion coefficients and Onsager coefficients for a system with $1/x = 0.25$, hard-core interactions, and volume conservation with $U_{\mathrm{CV}} = 1000$ and $v_0/v_- = 0.67$.

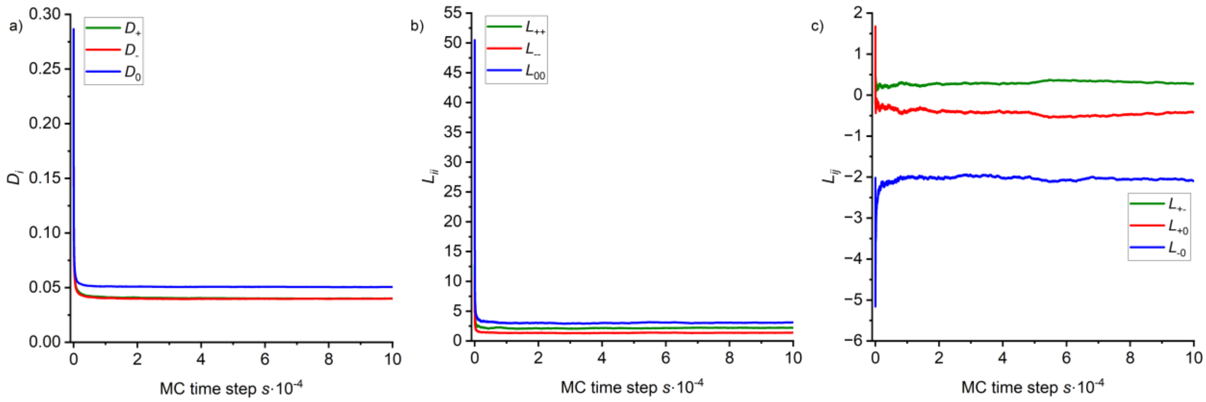

Fig. S3: Time-dependent diffusion coefficients and Onsager coefficients for a system with $1/x = 0.25$, hard-core interactions, and volume conservation with $U_{\mathrm{CV}} = 1000$ and $v_0/v_{-} = 0.67$, and Coulomb interactions with $U_{\mathrm{Coul}} = 5$.

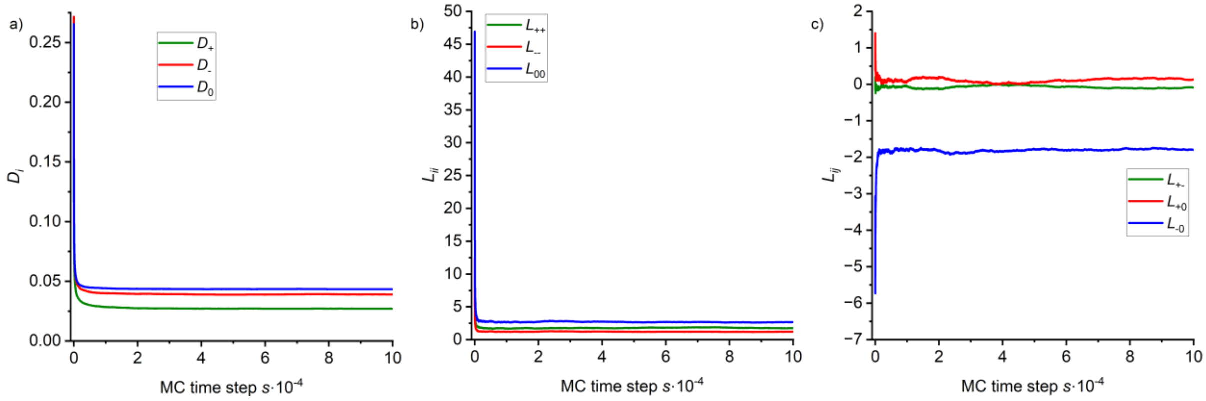

Fig. S4: Time-dependent diffusion coefficients and Onsager coefficients for a system with $1/x = 0.25$, hard-core interactions, and volume conservation with $U_{\mathrm{CV}} = 1000$ and $v_0/v_{-} = 0.67$, Coulomb interactions with $U_{\mathrm{Coul}} = 5$, and cation-solvent interactions with $U_{\mathrm{cat-sol}} = -2.5$.

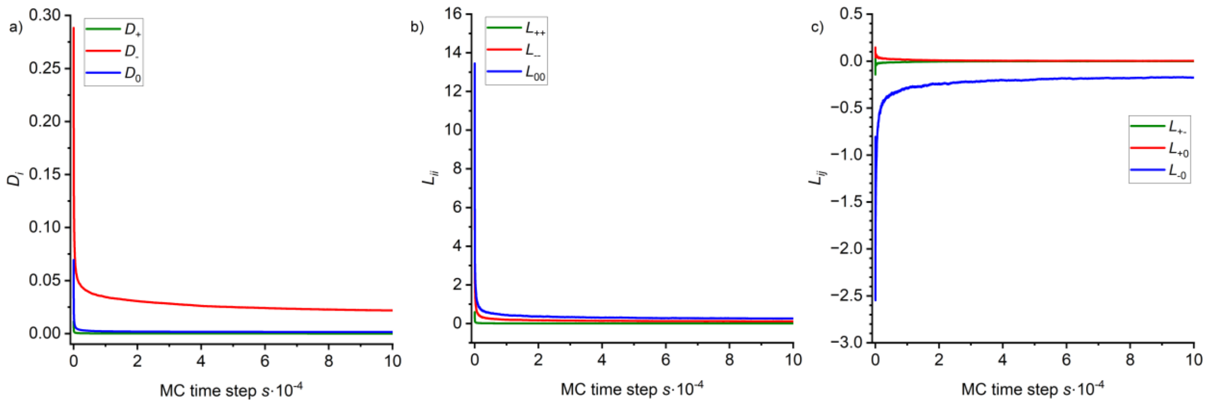

Fig. S5: Time-dependent diffusion coefficients and Onsager coefficients for a system with $1/x = 0.25$, hard-core interactions, and volume conservation with $U_{\mathrm{CV}} = 1000$ and $v_0/v_{-} = 0.67$, Coulomb interactions with $U_{\mathrm{Coul}} = 5$, and cation-solvent interactions with $U_{\mathrm{cat-sol}} = -7.0$.

Self and distinct parts of Onsager coefficients and jump rates of particles

Tab. S1: Self parts of Onsager coefficients for systems with only hard-core interactions.

$\frac{1}{x}$	L_{+}^{self}	L_{-}^{self}	L_{0}^{self}
0.1	1.40 ± 0.01	1.39 ± 0.01	13.99 ± 0.03
0.25	2.82 ± 0.01	2.80 ± 0.02	11.23 ± 0.03
0.5	4.18 ± 0.02	4.20 ± 0.02	8.38 ± 0.03
1.0	5.60 ± 0.01	5.64 ± 0.02	5.59 ± 0.02

Tab. S2: Distinct parts of Onsager coefficients and L_{-0} for systems with only hard-core interactions.

$\frac{1}{x}$	L_{++}^{distinct}	L_{--}^{distinct}	L_{00}^{distinct}	L_{-0}
0.1	0.38 ± 0.08	0.39 ± 0.05	41 ± 2	3.9 ± 0.3
0.25	1.4 ± 0.2	1.7 ± 0.2	27 ± 2	6.8 ± 0.5
0.5	3.9 ± 0.4	3.9 ± 0.2	15 ± 1	7.7 ± 0.5
1.0	5.8 ± 0.5	6.1 ± 0.3	5.9 ± 0.4	6.2 ± 0.3

Tab. S3: Jump rates of particles for systems with only hard-core interactions.

$\frac{1}{x}$	jump rate of the cations	jump rate of the anions	jump rate of the solvent
0.1	0.33 ± 0.00	0.33 ± 0.00	0.33 ± 0.00
0.25	0.33 ± 0.00	0.33 ± 0.00	0.33 ± 0.00
0.5	0.33 ± 0.00	0.33 ± 0.00	0.33 ± 0.00
1.0	0.33 ± 0.00	0.33 ± 0.00	0.33 ± 0.00

Tab. S4: Self parts of Onsager coefficients for systems with hard-core interactions and volume conservation with $U_{\text{CV}} = 1000$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	L_{+}^{self}	L_{-}^{self}	L_{0}^{self}
0.67	0.1	1.31 ± 0.01	1.27 ± 0.01	12.87 ± 0.01
	0.25	2.65 ± 0.01	2.54 ± 0.01	10.26 ± 0.03
	0.5	3.96 ± 0.02	3.79 ± 0.02	7.68 ± 0.02
	1.0	5.26 ± 0.02	5.02 ± 0.02	5.12 ± 0.02
0.90	0.1	1.32 ± 0.01	1.28 ± 0.01	12.83 ± 0.02
	0.25	2.64 ± 0.02	2.56 ± 0.01	10.24 ± 0.02
	0.5	3.93 ± 0.02	3.83 ± 0.02	7.67 ± 0.02
	1.0	5.29 ± 0.01	5.06 ± 0.02	5.08 ± 0.02
1.05	0.1	1.31 ± 0.01	1.27 ± 0.01	12.85 ± 0.03
	0.25	2.66 ± 0.02	2.58 ± 0.01	10.23 ± 0.03
	0.5	3.96 ± 0.01	3.81 ± 0.01	7.64 ± 0.02
	1.0	5.29 ± 0.02	5.07 ± 0.01	5.08 ± 0.01

Tab. S5: Distinct parts of Onsager coefficients and L_{-0} for systems with hard-core interactions and volume conservation with $U_{\mathrm{CV}} = 1000$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	$L_{++}^{\mathrm{distinct}}$	$L_{--}^{\mathrm{distinct}}$	$L_{00}^{\mathrm{distinct}}$	L_{-0}
0.67	0.1	0.20 ± 0.04	-0.26 ± 0.03	-10.61 ± 0.07	-1.51 ± 0.05
	0.25	0.6 ± 0.2	-0.88 ± 0.06	-6.6 ± 0.1	-2.47 ± 0.09
	0.5	1.2 ± 0.2	-1.88 ± 0.05	-3.4 ± 0.1	-2.85 ± 0.07
	1.0	2.2 ± 0.3	-3.46 ± 0.05	-1.64 ± 0.09	-2.33 ± 0.06
0.90	0.1	0.08 ± 0.04	-0.15 ± 0.04	-11.45 ± 0.06	-1.25 ± 0.04
	0.25	0.35 ± 0.09	-0.56 ± 0.07	-7.77 ± 0.09	-2.23 ± 0.08
	0.5	1.0 ± 0.2	-1.30 ± 0.08	-4.6 ± 0.1	-2.80 ± 0.09
	1.0	1.6 ± 0.3	-2.84 ± 0.07	-2.35 ± 0.09	-2.46 ± 0.08
1.05	0.1	0.07 ± 0.04	-0.12 ± 0.04	-11.81 ± 0.05	-1.09 ± 0.04
	0.25	0.5 ± 0.1	-0.42 ± 0.09	-8.26 ± 0.08	-2.06 ± 0.08
	0.5	0.8 ± 0.2	-1.07 ± 0.08	-5.16 ± 0.07	-2.61 ± 0.07
	1.0	1.8 ± 0.3	-2.5 ± 0.1	-2.75 ± 0.09	-2.45 ± 0.09

Tab. S6: Jump rates of particles for systems for systems with hard-core-interactions and volume conservation with $U_{\mathrm{CV}} = 1000$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	jump rate of the cations	jump rate of the anions	jump rate of the solvent
0.67	0.1	0.33 ± 0.01	0.30 ± 0.01	0.31 ± 0.01
	0.25	0.33 ± 0.01	0.30 ± 0.01	0.31 ± 0.01
	0.5	0.33 ± 0.01	0.30 ± 0.01	0.31 ± 0.01
	1.0	0.33 ± 0.01	0.30 ± 0.01	0.31 ± 0.01
0.90	0.1	0.33 ± 0.01	0.31 ± 0.01	0.31 ± 0.01
	0.25	0.33 ± 0.01	0.31 ± 0.01	0.31 ± 0.01
	0.5	0.33 ± 0.01	0.30 ± 0.01	0.31 ± 0.01
	1.0	0.33 ± 0.01	0.30 ± 0.01	0.30 ± 0.01
1.05	0.1	0.33 ± 0.01	0.31 ± 0.01	0.31 ± 0.01
	0.25	0.33 ± 0.01	0.31 ± 0.01	0.31 ± 0.01
	0.5	0.33 ± 0.01	0.31 ± 0.01	0.30 ± 0.01
	1.0	0.33 ± 0.01	0.30 ± 0.01	0.30 ± 0.01

Tab. S7: Self parts of Onsager coefficients for systems with hard-core-interactions, volume conservation with $U_{\mathrm{CV}} = 1000$ and Coulomb interactions with $U_{\mathrm{Coul}} = 5$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	L_{+}^{self}	L_{-}^{self}	L_{0}^{self}
0.67	0.1	1.02 ± 0.01	0.99 ± 0.01	12.61 ± 0.01
	0.25	1.92 ± 0.01	1.91 ± 0.01	9.71 ± 0.02
	0.5	2.77 ± 0.01	2.73 ± 0.01	6.98 ± 0.02
	1.0	3.48 ± 0.01	3.43 ± 0.01	4.45 ± 0.02
0.90	0.1	1.02 ± 0.01	1.00 ± 0.01	12.51 ± 0.03
	0.25	1.94 ± 0.01	1.92 ± 0.01	9.66 ± 0.01
	0.5	2.77 ± 0.01	2.72 ± 0.01	7.00 ± 0.01
	1.0	3.50 ± 0.02	3.44 ± 0.01	4.42 ± 0.01
1.05	0.1	1.02 ± 0.01	1.01 ± 0.01	12.58 ± 0.03

	0.25	1.94 ± 0.01	1.93 ± 0.01	9.69 ± 0.04
	0.5	2.77 ± 0.02	2.75 ± 0.01	6.97 ± 0.02
	1.0	3.49 ± 0.01	3.44 ± 0.01	4.41 ± 0.02

Tab. S8: Distinct parts of Onsager coefficients and L_{-0} for systems with hard-core-interactions, volume conservation with $U_{\mathrm{CV}} = 1000$ and Coulomb interactions with $U_{\mathrm{Coul}} = 5$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	$L_{++}^{\mathrm{distinct}}$	$L_{--}^{\mathrm{distinct}}$	$L_{00}^{\mathrm{distinct}}$	L_{-0}
0.67	0.1	0.04 ± 0.03	-0.08 ± 0.04	-10.59 ± 0.08	-1.35 ± 0.06
	0.25	0.30 ± 0.07	-0.54 ± 0.05	-6.7 ± 0.1	-2.05 ± 0.07
	0.5	0.4 ± 0.1	-1.13 ± 0.07	-3.4 ± 0.2	-2.4 ± 0.1
	1.0	0.59 ± 0.07	-2.19 ± 0.05	-1.7 ± 0.1	-1.85 ± 0.08
0.90	0.1	0.11 ± 0.04	-0.04 ± 0.02	-11.33 ± 0.04	-1.06 ± 0.03
	0.25	0.23 ± 0.09	-0.35 ± 0.05	-7.72 ± 0.07	-1.74 ± 0.06
	0.5	0.56 ± 0.07	-0.87 ± 0.07	-4.72 ± 0.09	-2.05 ± 0.08
	1.0	1.1 ± 0.2	-1.70 ± 0.06	-2.28 ± 0.08	-1.93 ± 0.07
1.05	0.1	0.06 ± 0.03	-0.07 ± 0.04	-11.72 ± 0.05	-0.89 ± 0.04
	0.25	0.31 ± 0.07	-0.18 ± 0.08	-8.10 ± 0.08	-1.66 ± 0.07
	0.5	0.5 ± 0.1	-0.61 ± 0.04	-5.02 ± 0.04	-2.04 ± 0.04
	1.0	0.9 ± 0.1	-1.35 ± 0.07	-2.52 ± 0.07	-1.99 ± 0.07

Tab. S9: Jump rates of particles for systems with hard-core-interactions, volume conservation with $U_{\mathrm{CV}} = 1000$ and Coulomb interactions with $U_{\mathrm{Coul}} = 5$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	jump rate of the cations	jump rate of the anions	jump rate of the solvent
0.67	0.1	0.29 ± 0.01	0.28 ± 0.01	0.31 ± 0.01
	0.25	0.29 ± 0.01	0.27 ± 0.01	0.31 ± 0.01
	0.5	0.28 ± 0.01	0.27 ± 0.01	0.31 ± 0.01
	1.0	0.28 ± 0.01	0.27 ± 0.01	0.31 ± 0.01
0.90	0.1	0.29 ± 0.01	0.28 ± 0.01	0.31 ± 0.01
	0.25	0.29 ± 0.01	0.28 ± 0.01	0.31 ± 0.01
	0.5	0.28 ± 0.01	0.27 ± 0.01	0.31 ± 0.01
	1.0	0.28 ± 0.01	0.27 ± 0.01	0.31 ± 0.01
1.05	0.1	0.29 ± 0.01	0.29 ± 0.01	0.31 ± 0.01
	0.25	0.29 ± 0.01	0.28 ± 0.01	0.31 ± 0.01
	0.5	0.28 ± 0.01	0.28 ± 0.01	0.31 ± 0.01
	1.0	0.28 ± 0.01	0.27 ± 0.01	0.31 ± 0.01

Tab. S10: Self parts of Onsager coefficients for systems with hard-core interactions, volume conservation with $U_{\mathrm{CV}} = 1000$, Coulomb interactions with $U_{\mathrm{Coul}} = 5$, and cation-solvent interactions with $U_{\mathrm{cat-sol}} = -2.5$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	L_{+}^{self}	L_{-}^{self}	L_{0}^{self}
0.67	0.1	0.68 ± 0.01	0.98 ± 0.01	11.51 ± 0.03
	0.25	1.30 ± 0.01	1.88 ± 0.01	8.34 ± 0.02
	0.5	2.00 ± 0.01	2.63 ± 0.01	5.63 ± 0.02

	1.0	2.81 ± 0.01	3.33 ± 0.01	3.37 ± 0.02
0.90	0.1	0.67 ± 0.01	0.99 ± 0.01	11.55 ± 0.04
	0.25	1.31 ± 0.01	1.89 ± 0.01	8.36 ± 0.02
	0.5	2.02 ± 0.01	2.67 ± 0.01	5.60 ± 0.02
	1.0	2.78 ± 0.01	3.34 ± 0.01	3.34 ± 0.01
1.05	0.1	0.668 ± 0.006	1.012 ± 0.005	11.59 ± 0.03
	0.25	1.30 ± 0.01	1.89 ± 0.01	8.32 ± 0.01
	0.5	2.01 ± 0.01	2.66 ± 0.01	5.62 ± 0.01
	1.0	2.80 ± 0.01	3.35 ± 0.01	3.34 ± 0.01

Tab. S11: Distinct parts of Onsager coefficients and L_{-0} for systems with hard-core interactions, volume conservation with $U_{\mathrm{CV}} = 1000$, Coulomb interactions with $U_{\mathrm{Coul}} = 5$, and cation-solvent interactions with $U_{\mathrm{cat-sol}} = -2.5$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	$L_{++}^{\mathrm{distinct}}$	$L_{--}^{\mathrm{distinct}}$	$L_{00}^{\mathrm{distinct}}$	L_{-0}
0.67	0.1	0.16 ± 0.02	-0.11 ± 0.02	-9.57 ± 0.05	-1.30 ± 0.03
	0.25	0.51 ± 0.06	-0.69 ± 0.03	-5.68 ± 0.08	-1.78 ± 0.05
	0.5	0.68 ± 0.06	-1.42 ± 0.03	-2.92 ± 0.07	-1.81 ± 0.05
	1.0	0.7 ± 0.1	-2.36 ± 0.04	-1.21 ± 0.08	-1.44 ± 0.05
0.90	0.1	0.13 ± 0.02	-0.06 ± 0.04	-10.40 ± 0.06	-1.04 ± 0.04
	0.25	0.46 ± 0.06	-0.47 ± 0.04	-6.61 ± 0.06	-1.57 ± 0.05
	0.5	0.44 ± 0.08	-1.18 ± 0.06	-3.76 ± 0.07	-1.65 ± 0.06
	1.0	0.8 ± 0.1	-1.91 ± 0.07	-1.58 ± 0.08	-1.59 ± 0.07
1.05	0.1	0.16 ± 0.03	-0.08 ± 0.02	-10.75 ± 0.04	-0.88 ± 0.02
	0.25	0.45 ± 0.06	-0.37 ± 0.04	-6.94 ± 0.04	-1.45 ± 0.04
	0.5	0.68 ± 0.08	-0.94 ± 0.05	-4.06 ± 0.04	-1.64 ± 0.04
	1.0	1.0 ± 0.2	-1.70 ± 0.06	-1.84 ± 0.05	-1.58 ± 0.06

Tab. S12: Jump rates of particles for systems with hard-core interactions, volume conservation with $U_{\mathrm{CV}} = 1000$, Coulomb interactions with $U_{\mathrm{Coul}} = 5$, and cation-solvent interactions with $U_{\mathrm{cat-sol}} = -2.5$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	jump rate of the cations	jump rate of the anions	jump rate of the solvent
0.67	0.1	0.20 ± 0.01	0.29 ± 0.01	0.30 ± 0.01
	0.25	0.22 ± 0.01	0.29 ± 0.01	0.29 ± 0.01
	0.5	0.24 ± 0.01	0.29 ± 0.01	0.28 ± 0.01
	1.0	0.25 ± 0.01	0.29 ± 0.01	0.27 ± 0.01
0.90	0.1	0.20 ± 0.01	0.30 ± 0.01	0.30 ± 0.01
	0.25	0.22 ± 0.01	0.30 ± 0.01	0.29 ± 0.01
	0.5	0.24 ± 0.01	0.29 ± 0.01	0.28 ± 0.01
	1.0	0.25 ± 0.01	0.29 ± 0.01	0.27 ± 0.01
1.05	0.1	0.20 ± 0.01	0.30 ± 0.01	0.30 ± 0.01
	0.25	0.22 ± 0.01	0.30 ± 0.01	0.29 ± 0.01
	0.5	0.24 ± 0.01	0.30 ± 0.01	0.28 ± 0.01
	1.0	0.25 ± 0.01	0.29 ± 0.01	0.26 ± 0.01

Tab. S13: Self parts of Onsager coefficients for systems with hard-core interactions, volume conservation with $U_{\mathrm{CV}} = 1000$, Coulomb interactions with $U_{\mathrm{Coul}} = 5$, and cation-solvent interactions with $U_{\mathrm{cat-sol}} = -7$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	L_+^{self}	L_-^{self}	L_0^{self}
0.67	0.1	0.00131 ± 0.00006	1.29 ± 0.02	4.86 ± 0.03
	0.25	0.00207 ± 0.00004	1.08 ± 0.01	0.295 ± 0.004
	0.5	0.0035 ± 0.0001	0.95 ± 0.01	0.0230 ± 0.0005
	1.0	0.0185 ± 0.0004	1.12 ± 0.01	0.0065 ± 0.0001
0.90	0.1	0.00118 ± 0.00005	1.32 ± 0.02	4.83 ± 0.01
	0.25	0.00208 ± 0.00003	1.12 ± 0.01	0.301 ± 0.004
	0.5	0.0035 ± 0.0001	0.98 ± 0.01	0.0229 ± 0.0005
	1.0	0.0187 ± 0.003	1.13 ± 0.01	0.0065 ± 0.0002
1.05	0.1	0.0013 ± 0.0001	1.33 ± 0.01	4.74 ± 0.02
	0.25	0.00210 ± 0.00004	1.13 ± 0.01	0.297 ± 0.004
	0.5	0.0035 ± 0.0001	0.99 ± 0.01	0.023 ± 0.001
	1.0	0.0196 ± 0.0004	1.15 ± 0.01	0.0068 ± 0.0001

Tab. S14: Distinct parts of Onsager coefficients and L_{-0} for systems with hard-core interactions, volume conservation with $U_{\mathrm{CV}} = 1000$, Coulomb interactions with $U_{\mathrm{Coul}} = 5$, and cation-solvent interactions with $U_{\mathrm{cat-sol}} = -7$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	$L_{++}^{\mathrm{distinct}}$	$L_{--}^{\mathrm{distinct}}$	$L_{00}^{\mathrm{distinct}}$	L_{-0}
0.67	0.1	0.0004 ± 0.0001	-0.42 ± 0.05	-2.9 ± 0.1	-1.30 ± 0.07
	0.25	0.0008 ± 0.0001	-0.96 ± 0.01	-0.03 ± 0.01	-0.18 ± 0.01
	0.5	0.0012 ± 0.0002	-0.93 ± 0.01	0.006 ± 0.001	-0.019 ± 0.001
	1.0	0.006 ± 0.001	-1.12 ± 0.01	0.0032 ± 0.0003	-0.0064 ± 0.0002
0.90	0.1	0.0005 ± 0.0002	-0.21 ± 0.04	-3.45 ± 0.05	-1.23 ± 0.04
	0.25	0.0006 ± 0.0001	-0.92 ± 0.02	-0.05 ± 0.01	-0.23 ± 0.01
	0.5	0.0013 ± 0.0003	-0.95 ± 0.01	0.003 ± 0.001	-0.023 ± 0.001
	1.0	0.005 ± 0.001	-1.12 ± 0.01	0.0027 ± 0.0004	-0.0082 ± 0.0003
1.05	0.1	0.0005 ± 0.0001	-0.20 ± 0.03	-3.77 ± 0.05	-1.02 ± 0.04
	0.25	0.0008 ± 0.0001	-0.78 ± 0.01	-0.07 ± 0.01	-0.24 ± 0.01
	0.5	0.0011 ± 0.0002	-0.97 ± 0.01	0.004 ± 0.001	-0.028 ± 0.001
	1.0	0.006 ± 0.001	-0.99 ± 0.01	0.0030 ± 0.0004	-0.0103 ± 0.0004

Tab. S15: Jump rates of particles for systems with hard-core interactions, volume conservation with $U_{\mathrm{CV}} = 1000$, Coulomb interactions with $U_{\mathrm{Coul}} = 5$, and cation-solvent interactions with $U_{\mathrm{cat-sol}} = -7$.

$\frac{v_0}{v_-}$	$\frac{1}{x}$	jump rate of the cations	jump rate of the anions	jump rate of the solvent
0.67	0.1	0.01 ± 0.01	0.40 ± 0.01	0.17 ± 0.01
	0.25	0.01 ± 0.01	0.31 ± 0.01	0.08 ± 0.01
	0.5	0.02 ± 0.01	0.25 ± 0.01	0.04 ± 0.01
	1.0	0.04 ± 0.01	0.22 ± 0.01	0.03 ± 0.01
0.90	0.1	0.01 ± 0.01	0.41 ± 0.01	0.17 ± 0.01
	0.25	0.01 ± 0.01	0.31 ± 0.01	0.07 ± 0.01

	0.5	0.02 ± 0.01	0.25 ± 0.01	0.04 ± 0.01
	1.0	0.04 ± 0.01	0.22 ± 0.01	0.03 ± 0.01
1.05	0.1	0.01 ± 0.01	0.41 ± 0.01	0.19 ± 0.01
	0.25	0.01 ± 0.01	0.31 ± 0.01	0.07 ± 0.01
	0.5	0.02 ± 0.01	0.25 ± 0.01	0.04 ± 0.01
	1.0	0.04 ± 0.01	0.22 ± 0.01	0.03 ± 0.01

Composition-dependent Onsager coefficients for systems with $\frac{v_0}{v_-} = 0.67$

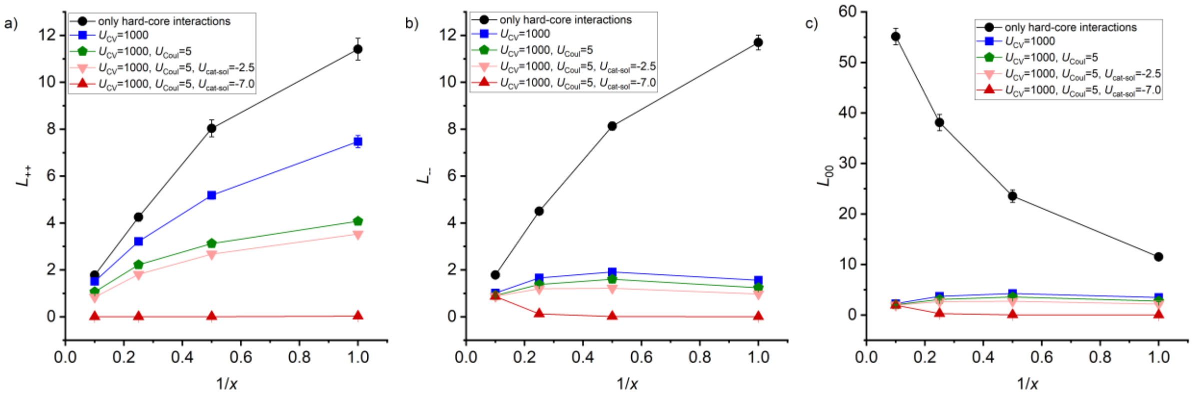

Fig. S6: Plots of the Onsager coefficients L_{++}, L_{--} and L_{00} vs. molar ratio $1/x$ for different energy parameters.

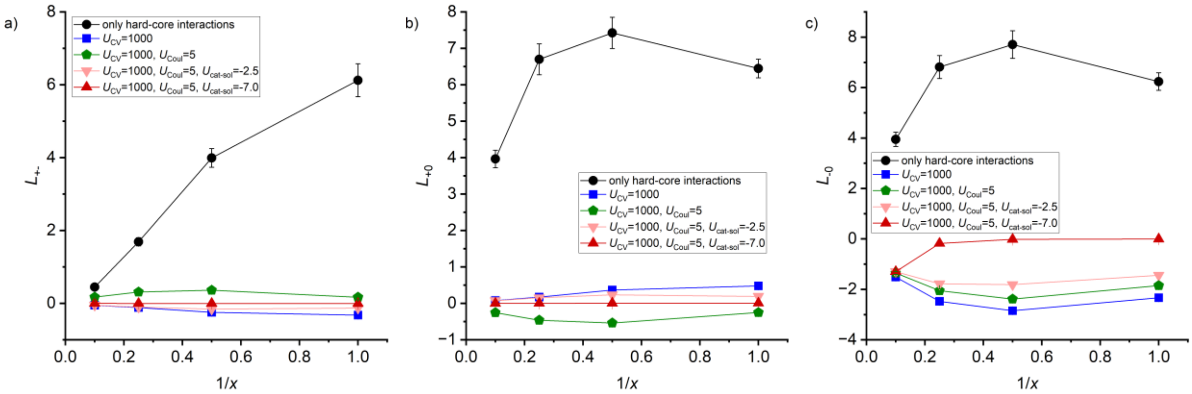

Fig. S7: Plots of the Onsager coefficients L_{+-}, L_{+0} and L_{-0} vs. molar ratio $1/x$ for different energy parameters.

Composition-dependent diffusion coefficients and correlation parameters for systems with $\frac{v_0}{v_-} = 0.90$

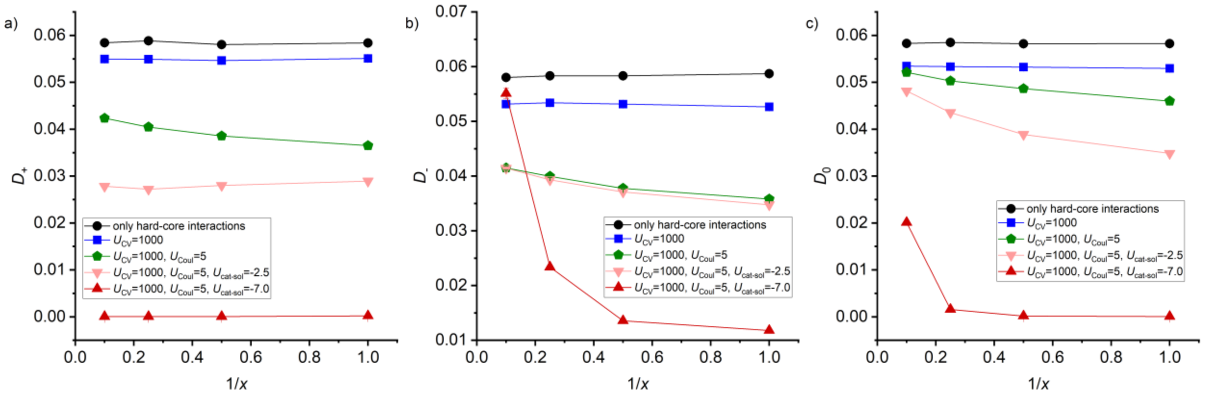

Fig. S8: Plots of the diffusion coefficients of D_+, D_-, and D_0 vs. the molar ratio $1/x$ for different energy parameters.

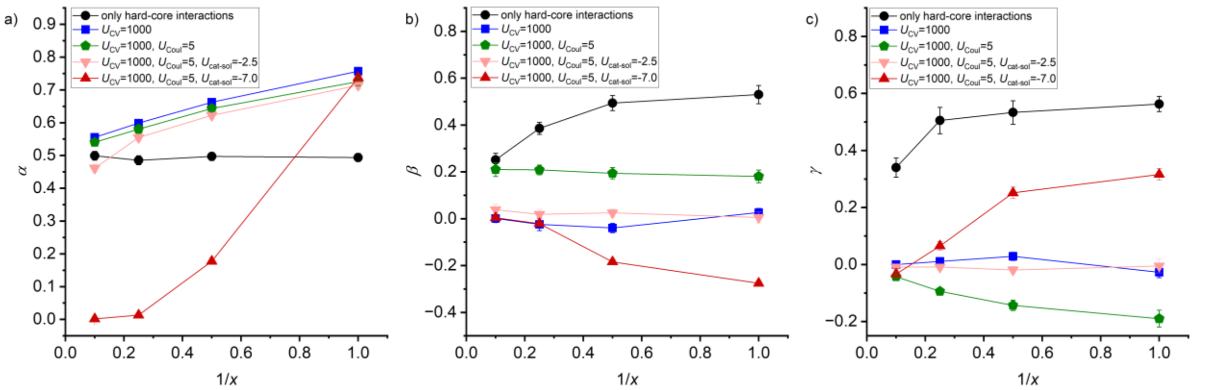

Fig. S9: Plots of correlation parameters α, β and γ vs. the molar ratio $1/x$ for different energy parameters.

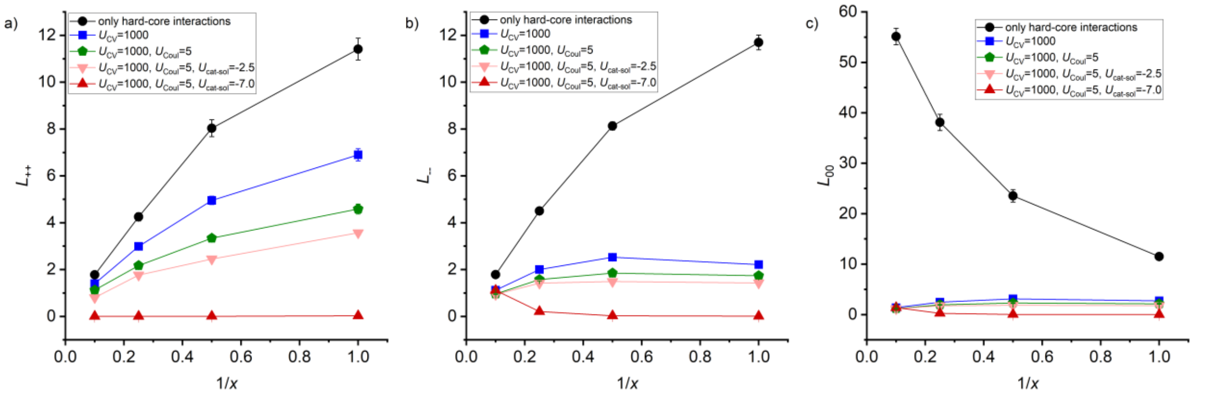

Fig. S10: Plots of the Onsager coefficients L_{++} , L_{--}, and L_{00} vs. the molar ratio $1/x$ for different energy parameters.

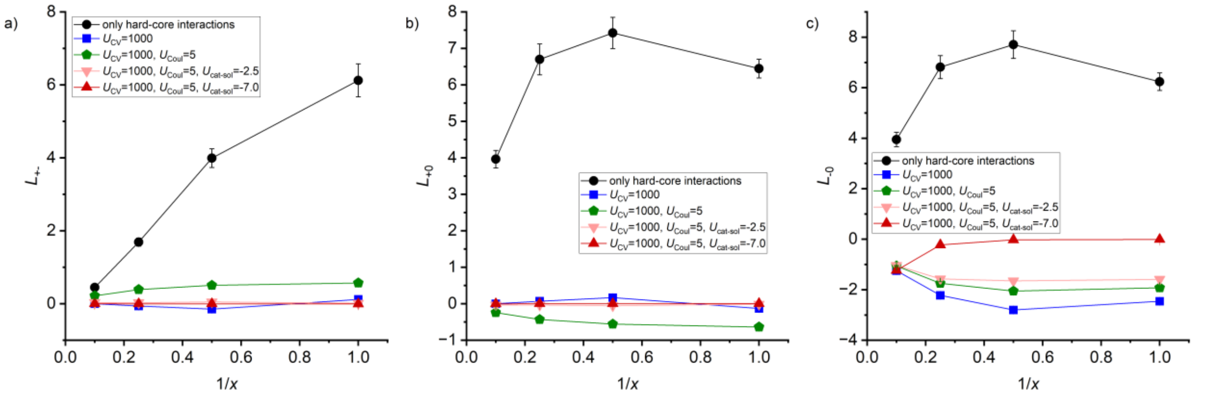

Fig. S11: Plots of the Onsager coefficients L_{+-} , L_{+0}, and L_{-0} vs. the molar ratio $1/x$ for different energy parameters.

Composition-dependent diffusion coefficients and correlation parameters for systems with $\frac{v_0}{v_-} = 1.05$

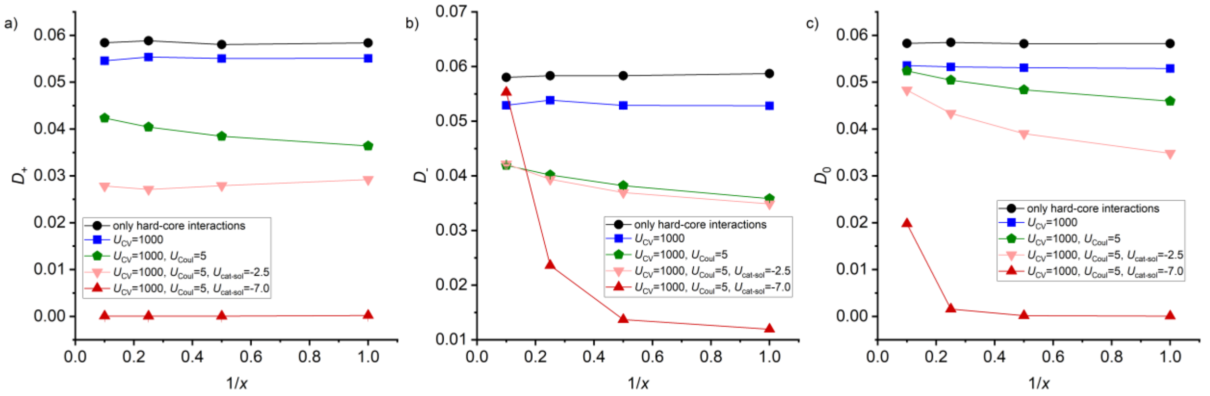

Fig. S12: Plots of the diffusion coefficients of D_+, D_-, and D_0 vs. the molar ratio $1/x$ for different energy parameters.

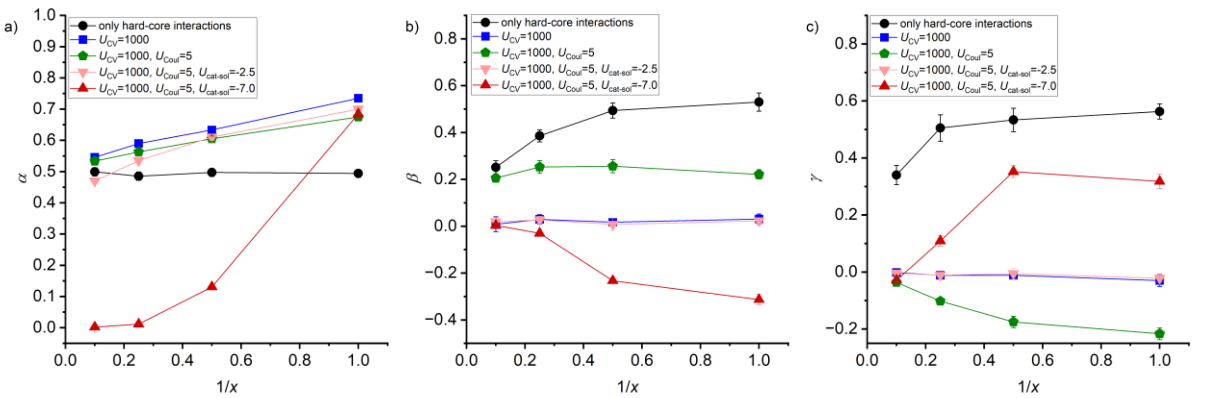

Fig. S13: Plots of correlation parameters α, β and γ vs. the molar ratio $1/x$ for different energy parameters.

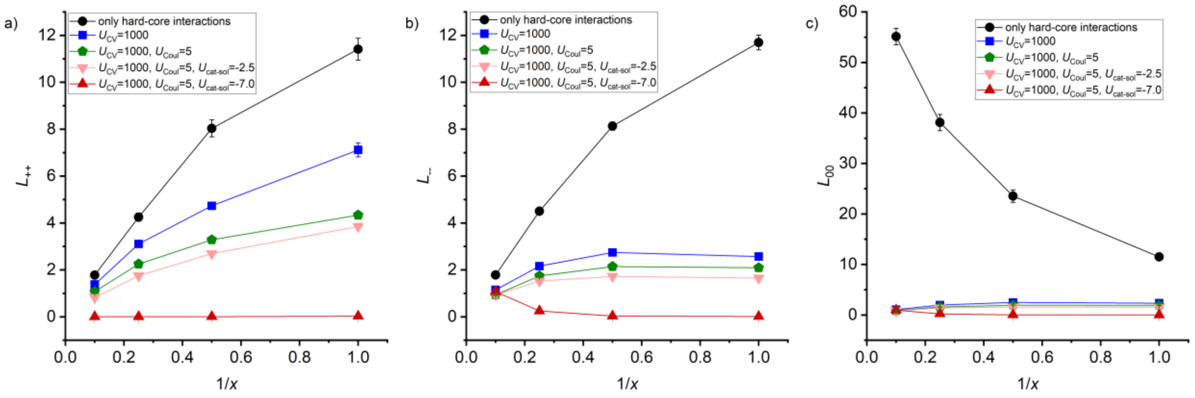

Fig. S14: Plots of the Onsager coefficients L_{++}, L_{--}, and L_{00} vs. the molar ratio $1/x$ for different energy parameters.

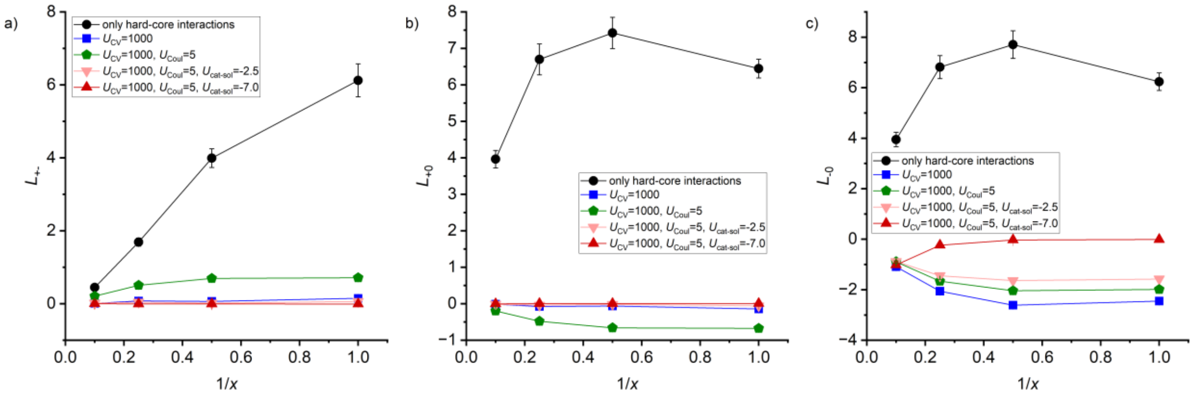

Fig. S15: Plots of the Onsager coefficients L_{+-}, L_{+0}, and L_{-0} vs. the molar ratio $1/x$ for different energy parameters.

Simulation results for different energy penalty parameter for center-of-volume shifts U_{CV}

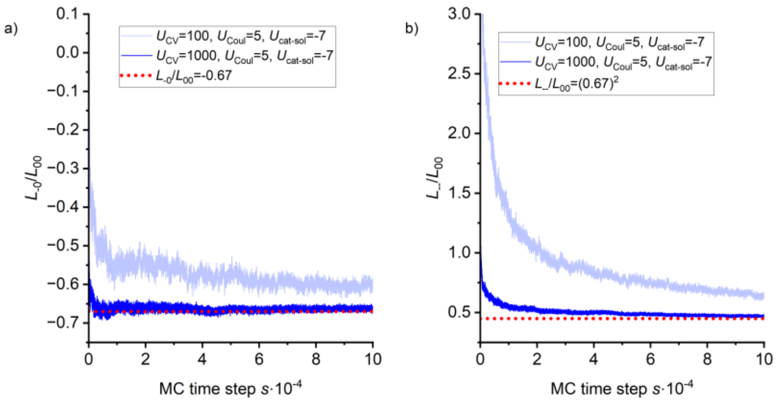

Fig. S16: Time-dependent ratio of the Onsager coefficients (a) L_{-0}/L_{00} and (b) L_{--}/L_{00} for a system with a molar ratio of $1/x = 0.5$ and with different values of U_{CV}. For comparison, theoretical results for strict volume conservation are shown as dotted lines. Volume conservation is considered as sufficiently strict, if the mean value of the ratios L_{-0}/L_{00} and (b)

L_{--}/L_{00} deviate by less than 5% from the predictions of Eq. (17) and (18) over the last $3 \cdot 10^4$ MC time steps of the equilibrium simulations.

Influence of the volume ratio v_0/v_- on the parameter α

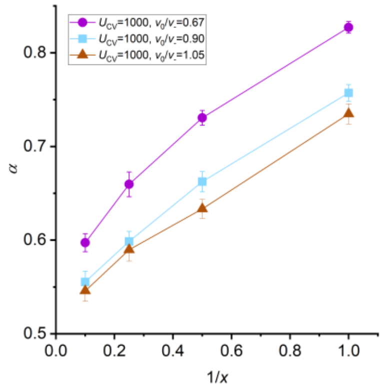

Fig. S17: Plot of the parameter α vs. the molar ratio $1/x$ for the systems with only volume conservation and different volume ratios v_0/v_-.

Derivation of Eq. (20) for analyzing the relative contributions of anions and solvent molecules to volume conservation

The sum over all displacement vectors of the anions is given by:

$$\sum_{i}^{N_-} \Delta\vec{r_i}(t) \quad \text{(S1)}$$

The sum over all displacement vectors of the solvent molecules is given by:

$$\sum_{j}^{x \cdot N_-} \Delta\vec{r_j}(t) \quad \text{(S2)}$$

In the case of strict volume conservation, the following equation must hold:

$$\frac{v_-}{v_0} \cdot \sum_{i}^{N_-} \Delta\vec{r_i}(t) = -\sum_{j}^{x \cdot N_-} \Delta\vec{r_j}(t) \quad \text{(S3)}$$

which can be rewritten as:

$$\frac{v_-}{v_0} \cdot \sum_{i}^{N_-} \Delta\vec{r_i}(t) + \sum_{j}^{x \cdot N_-} \Delta\vec{r_j}(t) = 0 \quad \text{(S4)}$$

In Monte Carlo units, the displacement vectors of the species i and j are linked to the transport coefficients L_{ij} by:

$$L_{ij} = \lim_{t\to\infty} \frac{d}{dt}\left[\left(\sum_{i}^{N_i} \Delta\vec{r_i}(t)\right)\left(\sum_{j}^{N_j} \Delta\vec{r_j}(t)\right)\right] \quad \text{(S5)}$$

Squaring Eq. (S4) yields:

$$\left(\frac{v_-}{v_0}\right)^2 \cdot \left[\left(\sum_i^{N_-} \Delta\vec{r_i}(t)\right)^2\right] + \left[\left(\sum_j^{x\cdot N_-} \Delta\vec{r_j}(t)\right)^2\right] - 2\cdot\frac{v_-}{v_0}\cdot\sum_i^{N_-}\Delta\vec{r_i}(t)\cdot\sum_j^{x\cdot N_-}\Delta\vec{r_j}(t) = 0 \tag{S6}$$

Taking into account Eq. (S5) results in:

$$\left(\frac{v_-}{v_0}\right)^2 \cdot L_{--} + L_{00} + 2\cdot\frac{v_-}{v_0}L_{-0} = 0 \tag{S7}$$

$$\frac{v_-}{v_0}\cdot L_{--} + \frac{v_0}{v_-}L_{00} + 2\cdot L_{-0} = 0 \tag{S8}$$

Next the Onsager coefficients L_{--} and L_{00} are split into self and distinct parts:

$$\frac{v_-}{v_0}\cdot L_-^{\text{self}} + \frac{v_0}{v_-}L_0^{\text{self}} + \frac{v_-}{v_0}\cdot L_{--}^{\text{distinct}} + \frac{v_0}{v_-}L_0^{\text{distinct}} + 2\cdot L_{-0} = 0 \tag{S9}$$

and the equation is divided by $\left(\frac{v_-}{v_0}\cdot L_-^{\text{self}} + \frac{v_0}{v_-}L_0^{\text{self}}\right)$:

$$0 = \frac{\frac{v_-}{v_0}\cdot L_-^{\text{self}} + \frac{v_0}{v_-}L_0^{\text{self}}}{\frac{v_-}{v_0}\cdot L_-^{\text{self}} + \frac{v_0}{v_-}L_0^{\text{self}}} + \frac{\frac{v_-}{v_0}\cdot L_{--}^{\text{distinct}}}{\frac{v_-}{v_0}\cdot L_-^{\text{self}} + \frac{v_0}{v_-}L_0^{\text{self}}} + \frac{\frac{v_0}{v_-}L_{00}^{\text{distinct}}}{\frac{v_-}{v_0}\cdot L_-^{\text{self}} + \frac{v_0}{v_-}L_0^{\text{self}}} + \frac{2\cdot L_{-0}}{\frac{v_-}{v_0}\cdot L_-^{\text{self}} + \frac{v_0}{v_-}L_0^{\text{self}}} \tag{S10}$$

$$0 = 1 + \frac{L_{--}^{\text{distinct}}}{L_-^{\text{self}} + \left(\frac{v_0}{v_-}\right)^2 L_0^{\text{self}}} + \frac{\left(\frac{v_0}{v_-}\right)^2 L_{00}^{\text{distinct}}}{L_-^{\text{self}} + \left(\frac{v_0}{v_-}\right)^2 L_0^{\text{self}}} + \frac{2\frac{v_0}{v_-}\cdot L_{-0}}{L_-^{\text{self}} + \left(\frac{v_0}{v_-}\right)^2 L_0^{\text{self}}} \tag{S11}$$

Finally, we obtain:

$$-1 = \frac{L_{--}^{\text{distinct}}}{L_-^{\text{self}} + \left(\frac{v_0}{v_-}\right)^2 L_0^{\text{self}}} + \frac{\left(\frac{v_0}{v_-}\right)^2 L_{00}^{\text{distinct}}}{L_-^{\text{self}} + \left(\frac{v_0}{v_-}\right)^2 L_0^{\text{self}}} + \frac{2\frac{v_0}{v_-}\cdot L_{-0}}{L_-^{\text{self}} + \left(\frac{v_0}{v_-}\right)^2 L_0^{\text{self}}} \tag{20}$$

Relative contributions to the volume conservation for systems with $\frac{v_0}{v_-} = 0.9$

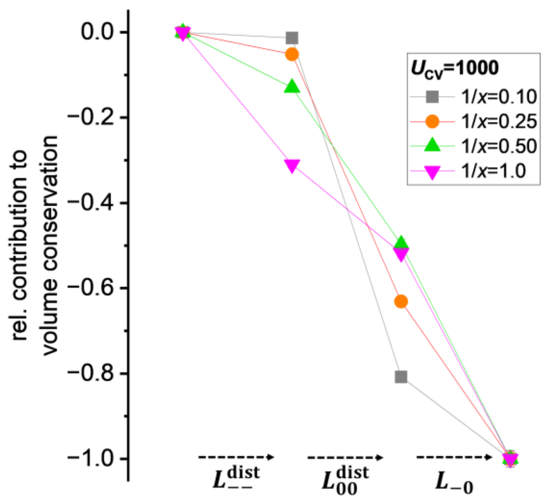

Fig. S18: Relative contributions of L_{--}^{dist}, L_{00}^{dist} and L_{-0} to volume conservation for different molar ratios $1/x$ in systems with hard-core interactions between the particles and with strict volume conservation ($U_{\text{CV}} = 1000$) at a volume ratio of v_0/v_-=0.90.

Derivation of Eq. (23) for the ratio L_{salt} / L_{ion}

The combination of equations (23) and (24) yields:

$$\frac{L_{\text{salt}}}{L_{\text{ion}}} = \frac{L_{++} \cdot L_{--} - L_{+-}^2}{(L_{++} + L_{--} - 2L_{+-})^2} \tag{S12}$$

Dividing all Onsager coefficients in Eq. (S12) by $(L_{++} + L_{-})$ and taking into account that

$$\alpha = \frac{L_{++}}{L_{++} + L_{-}} \tag{14}$$

$$1 - \alpha = \frac{L_{--}}{L_{++} + L_{-}} \tag{S13}$$

$$\beta = \frac{2 \cdot L_{+-}}{L_{++} + L_{-}} \tag{15}$$

results in:

$$\frac{L_{\text{salt}}}{L_{\text{ion}}} = \frac{\alpha(1-\alpha) - (0.5 \cdot \beta)^2}{(1-\beta)^2} \tag{23}$$

Comparison of two different cutoffs for the attractive cation-solvent interaction for systems with $\frac{v_0}{v_-} = 0.67$

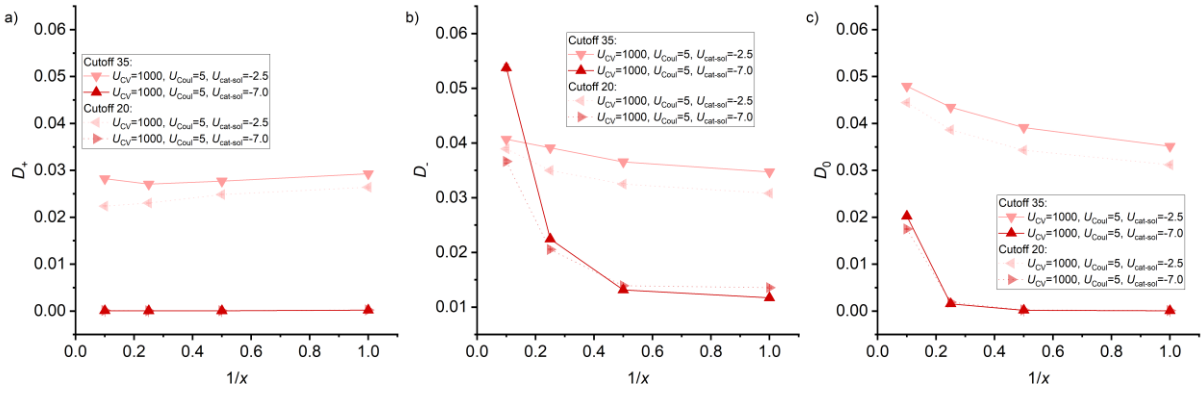

Fig. S19: Plots of the diffusion coefficients of D_+, D_-, and D_0 vs. the molar ratio $1/x$ for different cutoffs for the attractive cation-solvent interaction.

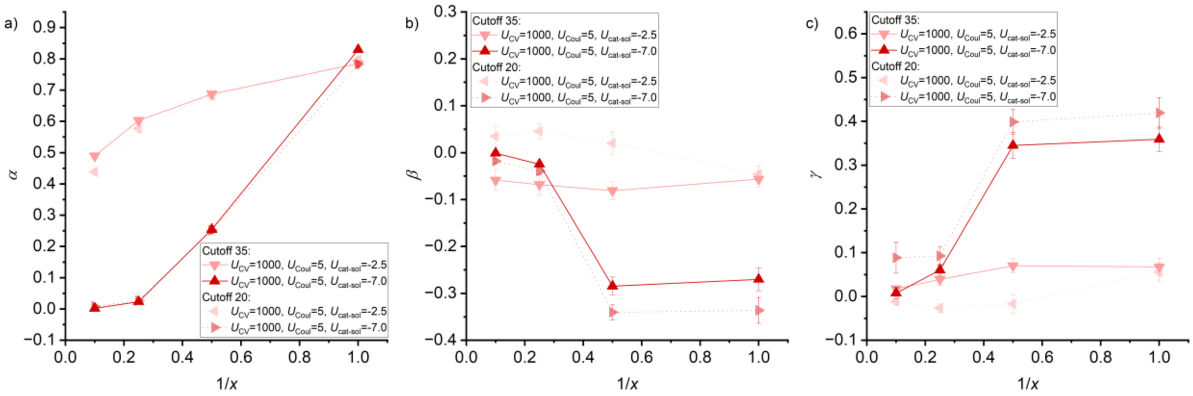

Fig. S20: Plots of correlation parameters α, β and γ vs. the molar ratio $1/x$ for different cutoffs for the attractive cation-solvent interaction.

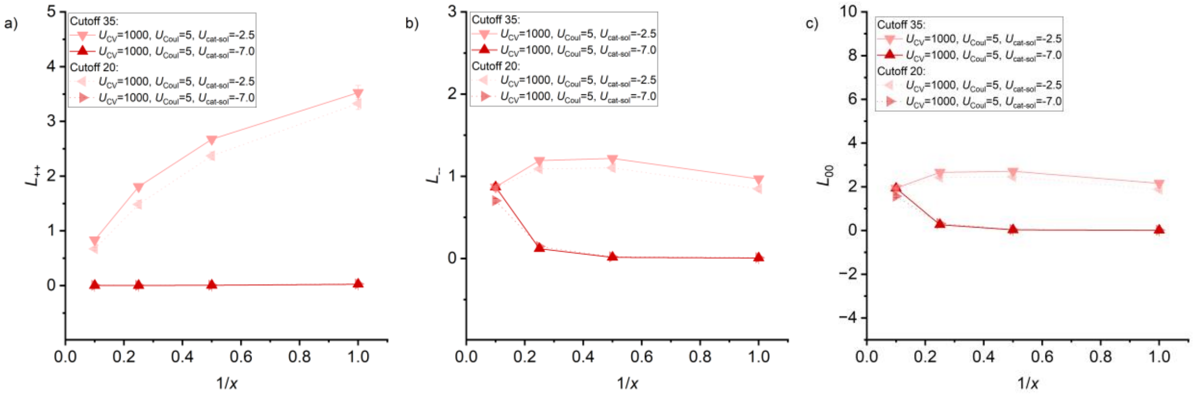

Fig. S21: Plots of the Onsager coefficients L_{++}, L_{--}, and L_{00} vs. the molar ratio $1/x$ for different cutoffs for the attractive cation-solvent interaction.

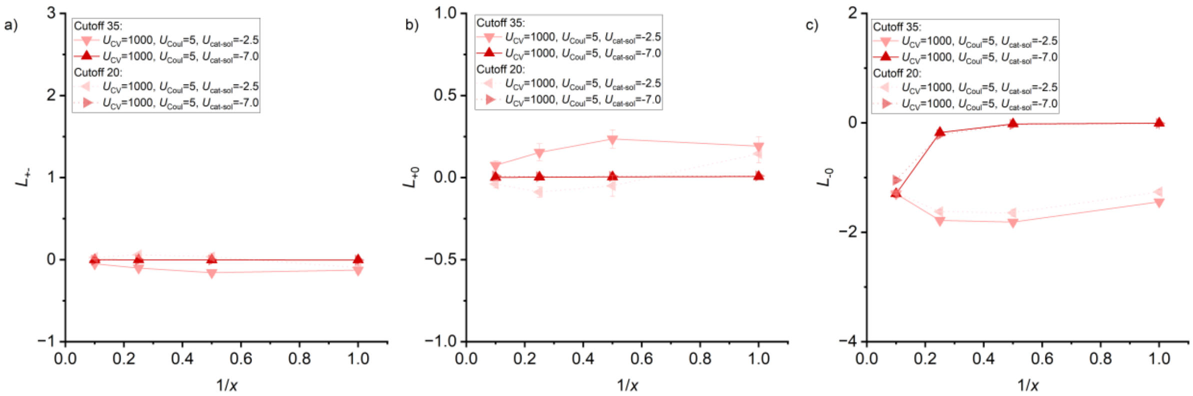

Fig. S22: Plots of the Onsager coefficients L_{+-}, L_{+0}, and L_{-0} vs. the molar ratio $1/x$ for different cutoffs for the attractive cation-solvent interaction.

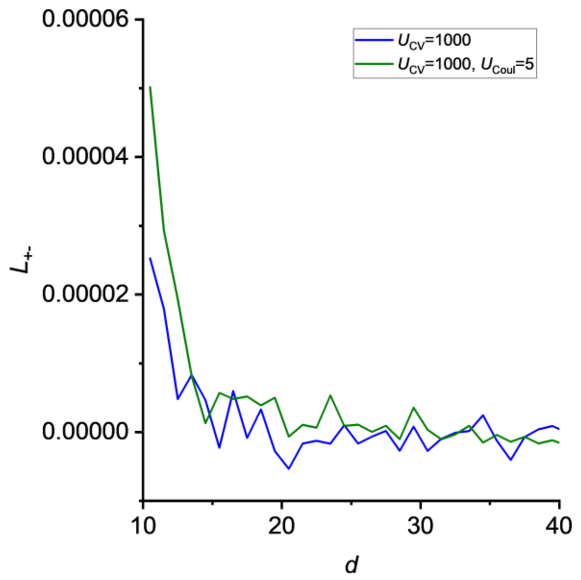

Fig. S23: Dependence of the Onsager coefficient L_{+-} on the initial cation-anion distance *d* for two systems with a molar ratio of $1/x = 1$, $\frac{v_0}{v_-} = 0.67$ and with strict volume conservation ($U_{\mathrm{CV}} = 1000$). Coulomb interactions with $U_{\mathrm{Coul}} = 5$ lead to enhanced positive cation-anion correlations when the initial separation distance of cations and anion is small.

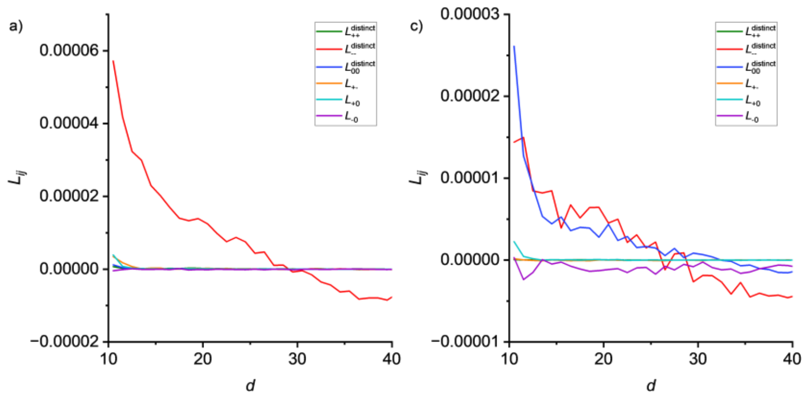

Fig. S24: Dependence of Onsager coefficients $L_{++}^{\mathrm{distinct}}$, $L_{--}^{\mathrm{distinct}}$, $L_{00}^{\mathrm{distinct}}$ L_{+-} , L_{+0}, and L_{-0} on the initial distance between the respective particles, *d,* for systems with a molar ratio of a) $1/x = 1$ and of b) $1/x = 0.25$ and $\frac{v_0}{v_-} = 0.67$ with hard-core interactions, volume conservation with $U_{\mathrm{CV}} = 1000$, Coulomb interactions with $U_{\mathrm{Coul}} = 5$, and cation-solvent interactions with $U_{\mathrm{cat-sol}} = -7$.