Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting information for

Role of density and electrostatic interactions on the viscosity and non-newtonian behavior of ionic liquid - a molecular dynamics study

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1. Block average data for viscosity error estimation

Table S1 – Viscosity data η_i from each block *i* used to estimate the error of computed viscosity from NEMD simulations of system S1 (electrostatic interactions attenuated by a relative dielectric constant of 5 and density equilibrated at 1 atm). Values in mPa.s.

γ'(GHz)	η_1	η_2	η_3	η_4	η_5	η_{total}	error
0.2	8.031	8.188	7.860	7.247	7.850	7.835	0.3568
0.3	7.438	7.814	7.872	8.148	7.710	7.796	0.2576
0.5	8.224	7.938	8.287	8.192	8.147	8.158	0.1329
0.8	8.047	7.835	7.993	7.809	7.775	7.892	0.1204
1.0	7.656	7.775	7.679	7.577	7.713	7.680	0.0728
2.0	6.983	7.045	7.016	7.003	7.062	7.022	0.0316
3.0	6.375	6.452	6.394	6.403	6.396	6.404	0.0286
5.0	5.399	5.357	5.399	5.382	5.382	5.384	0.0173
10.0	3.963	3.956	3.969	3.943	3.953	3.957	0.0101
20.0	2.770	2.765	2.761	2.766	2.774	2.767	0.0050
30.0	2.221	2.219	2.219	2.221	2.219	2.220	0.0009
40.0	1.894	1.893	1.895	1.894	1.895	1.894	0.0007
50.0	1.679	1.676	1.677	1.677	1.678	1.677	0.0014
100.0	1.159	1.159	1.158	1.158	1.158	1.158	0.0004
200.0	0.822	0.822	0.822	0.822	0.823	0.822	0.0005

Table S2 – Viscosity data η_i from each block *i* used to estimate the error of computed viscosity from NEMD simulations of system S2 (electrostatic interactions attenuated by a relative dielectric constant of 15 and density equilibrated at 1 atm). Values in mPa.s.

γ'(GHz)	η_1	η_2	η_3	η_4	η_5	η_{total}	error
0.3	1.557	1.581	1.486	1.595	1.602	1.564	0.0469
0.5	1.468	1.474	1.566	1.504	1.465	1.496	0.0424
0.8	1.586	1.418	1.457	1.579	1.525	1.513	0.0743
1.0	1.533	1.544	1.507	1.516	1.441	1.508	0.0402
2.0	1.467	1.475	1.482	1.499	1.475	1.480	0.0121
3.0	1.474	1.486	1.514	1.490	1.476	1.488	0.0161
5.0	1.469	1.461	1.462	1.464	1.474	1.466	0.0051
10.0	1.406	1.412	1.405	1.400	1.412	1.407	0.0053
20.0	1.272	1.275	1.272	1.277	1.276	1.274	0.0022
30.0	1.159	1.157	1.157	1.156	1.160	1.158	0.0015
40.0	1.063	1.064	1.066	1.064	1.063	1.064	0.0011
50.0	0.989	0.989	0.988	0.989	0.989	0.989	0.0003
100.0	0.767	0.767	0.766	0.767	0.766	0.767	0.0004
200.0	0.587	0.587	0.587	0.587	0.587	0.587	0.0002

Table S3 – Viscosity data η_i from each block *i* used to estimate the error of computed viscosity from NEMD simulations of system S3 (without charges and equilibrated at 1 atm). Values in mPa.s.

γ'(GHz)	η_1	η_2	η_3	η_4	η_5	η_{total}	error
0.5	0.685	0.713	0.701	0.797	0.762	0.732	0.0466
0.8	0.720	0.722	0.728	0.736	0.685	0.718	0.0195
1.0	0.745	0.741	0.734	0.738	0.721	0.736	0.0091
2.0	0.744	0.721	0.727	0.693	0.728	0.722	0.0184
3.0	0.710	0.766	0.743	0.723	0.728	0.734	0.0216
5.0	0.730	0.718	0.724	0.732	0.733	0.727	0.0062
10.0	0.716	0.729	0.725	0.719	0.725	0.723	0.0053
20.0	0.710	0.709	0.707	0.710	0.709	0.709	0.0010
30.0	0.687	0.687	0.689	0.689	0.687	0.688	0.0010
40.0	0.664	0.665	0.665	0.665	0.664	0.665	0.0005
50.0	0.642	0.641	0.641	0.642	0.642	0.642	0.0005
100.0	0.552	0.552	0.552	0.553	0.553	0.552	0.0002
200.0	0.455	0.455	0.455	0.455	0.455	0.455	0.0001

Table S4 – Viscosity data η_i from each block *i* used to estimate the error of computed viscosity from NEMD simulations of system S4 (without charges and with the same density of S1). Values in mPa.s.

γ'(GHz)	η_1	η_2	η_3	η_4	η_5	η_{total}	error
0.2	1.480	1.657	1.626	1.957	1.481	1.640	0.1947
0.3	1.695	1.760	1.559	1.707	1.539	1.652	0.0973
0.5	1.755	1.799	1.659	1.724	1.739	1.735	0.0508
0.8	1.654	1.604	1.728	1.640	1.641	1.653	0.0455
1.0	1.661	1.678	1.617	1.666	1.615	1.647	0.0293
2.0	1.660	1.668	1.695	1.665	1.663	1.670	0.0141
3.0	1.666	1.646	1.681	1.681	1.656	1.666	0.0153
5.0	1.672	1.643	1.670	1.658	1.668	1.662	0.0121
10.0	1.603	1.603	1.602	1.595	1.602	1.601	0.0036
20.0	1.451	1.453	1.452	1.454	1.454	1.453	0.0011
30.0	1.322	1.320	1.318	1.319	1.320	1.320	0.0017
40.0	1.214	1.213	1.215	1.212	1.212	1.213	0.0013
50.0	1.128	1.128	1.127	1.127	1.129	1.128	0.0007
100.0	0.871	0.870	0.871	0.871	0.870	0.871	0.0003
200.0	0.665	0.665	0.665	0.665	0.665	0.665	0.0001

Table S5 – Viscosity data η_i from each block *i* used to estimate the error of computed viscosity from NEMD simulations of system S5 (without charges and with the same density of S2). Values in mPa.s.

γ'(GHz)	η_1	η_2	η_3	η_4	η_5	η_{total}	error
0.3	1.063	1.103	0.795	1.019	0.966	0.989	0.1201
0.5	1.017	1.111	1.079	1.082	1.104	1.079	0.0370
0.8	0.946	1.053	0.991	0.968	0.967	0.985	0.0412
1.0	0.997	0.991	0.996	1.020	1.027	1.006	0.0162
2.0	1.009	1.010	1.021	1.010	0.974	1.005	0.0179
3.0	0.995	1.002	1.030	1.010	1.028	1.013	0.0154
5.0	1.002	0.994	0.997	1.009	0.996	1.000	0.0060
10.0	0.991	0.981	0.984	0.985	0.989	0.986	0.0040
20.0	0.945	0.948	0.942	0.946	0.948	0.946	0.0022
30.0	0.898	0.899	0.900	0.902	0.902	0.900	0.0017
40.0	0.854	0.855	0.855	0.855	0.855	0.855	0.0005
50.0	0.815	0.815	0.814	0.814	0.814	0.814	0.0006
100.0	0.671	0.671	0.671	0.670	0.671	0.671	0.0003
200.0	0.534	0.535	0.536	0.535	0.535	0.535	0.0005

2. Parameters of Carreau equation fit

Table S6 – Parameter λ (ns) for Carreau equation fit for the dependence of viscosity, dimer lifetime and relative differences of radial distribution function with the shear rate in the five systems studied

Property	S1	S2	S3	S4	S5
viscosity	0.372	0.059	0.022	0.050	0.033
E_{coul}	0.079	0.032	----	----	----
$E_{dispesion}$	0.049	0.018	0.011	0.021	0.014
$\Delta g(r)_{rel}$ cation-anion 1st	0.099	0.033	0.020	0.043	0.025
$\Delta g(r)_{rel}$ cation-anion 2nd	0.193	0.073	----	----	----
$\Delta g(r)_{rel}$ cation-anion 3rd	0.281	0.059	0.016	0.035	0.026
$\Delta g(r)_{rel}$ anion-anion	0.319	0.060	0.066	0.106	0.103
$\Delta g(r)_{rel}$ cation-cation	0.195	0.044	----	----	----
t_p cation-anion	0.478	0.102	0.052	0.102	0.065
t_p anion-anion	0.506	0.094	0.046	0.088	0.057
t_p cation-cation	0.508	0.116	0.069	0.111	0.082

Table S7 – Parameter *n* for Carreau equation fit for the dependence of viscosity, dimer lifetime and relative differences of radial distribution function with the shear rate in the five systems studied

Property	S1	S2	S3	S4	S5
viscosity	0.472	0.624	0.691	0.599	0.670
E_{coul}	0.985	0.970	----	----	----
$E_{dispesion}$	0.981	0.981	0.981	0.977	0.978
$\Delta g(r)_{rel}$ cation-anion 1st	0.926	0.880	0.842	0.867	0.850
$\Delta g(r)_{rel}$ cation-anion 2nd	0.657	0.600	----	----	----
$\Delta g(r)_{rel}$ cation-anion 3rd	0.492	-0.012	0.351	0.503	0.491
$\Delta g(r)_{rel}$ anion-anion	1.171	1.098	1.018	1.027	1.017
$\Delta g(r)_{rel}$ cation-cation	0.784	0.761	----	----	----
t_p cation-anion	0.294	0.376	0.453	0.379	0.414
t_p anion-anion	0.324	0.404	0.464	0.386	0.424
t_p cation-cation	0.265	0.394	0.543	0.420	0.502

3. Additional images with Carreau equation fit

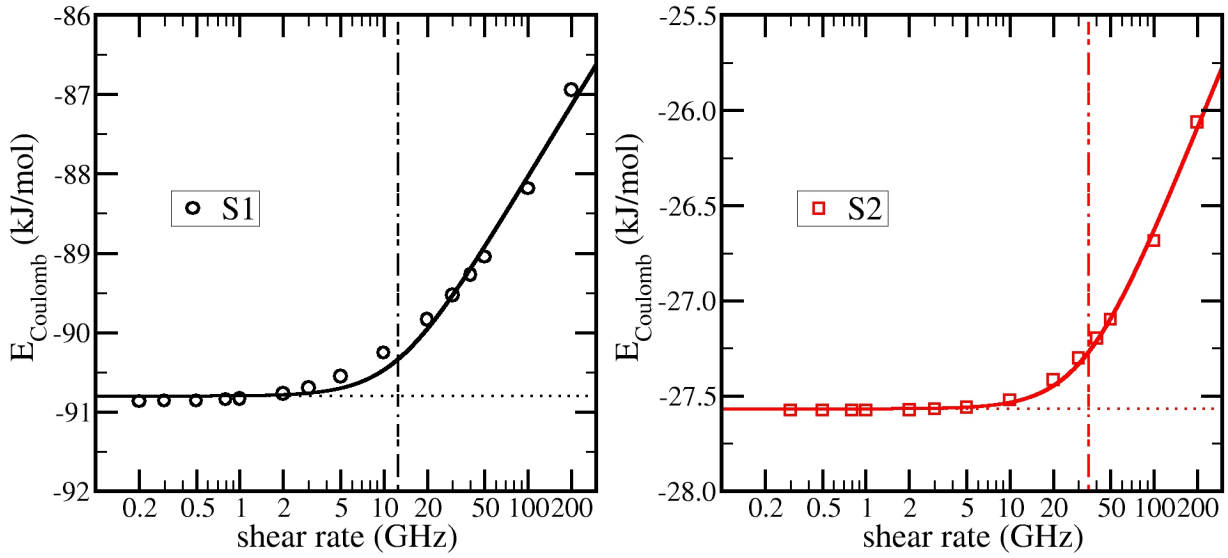

Figure S1 – Electrostatic energy per ion pair in systems S1 (left) and S2 (right) at different shear rates. The solid lines are the Carreau equation fit, the horizontal dashed lines represent the value at zero shear computed by the best fit and vertical dashed lines give the reciprocal of the parameter λ in GHz.

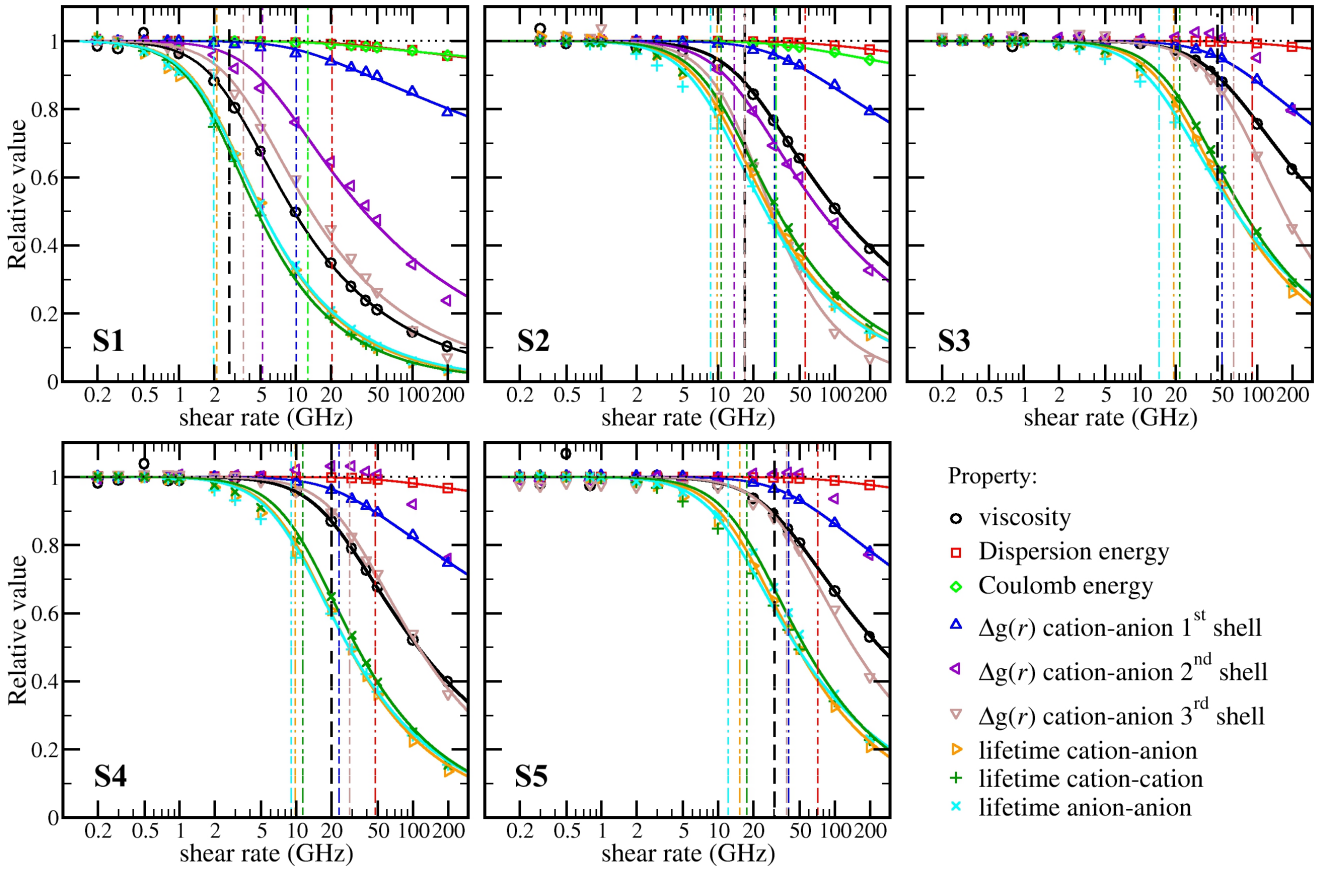

Figure S2 – Relative changes of several properties in relation to zero shear value with Carreau equation fit (solid lines with the same colors). Vertical dashed lines indicates the values of λ^{-1} (GHz) for each fit.

4. Cation-cation and anion-anion radial distribution function

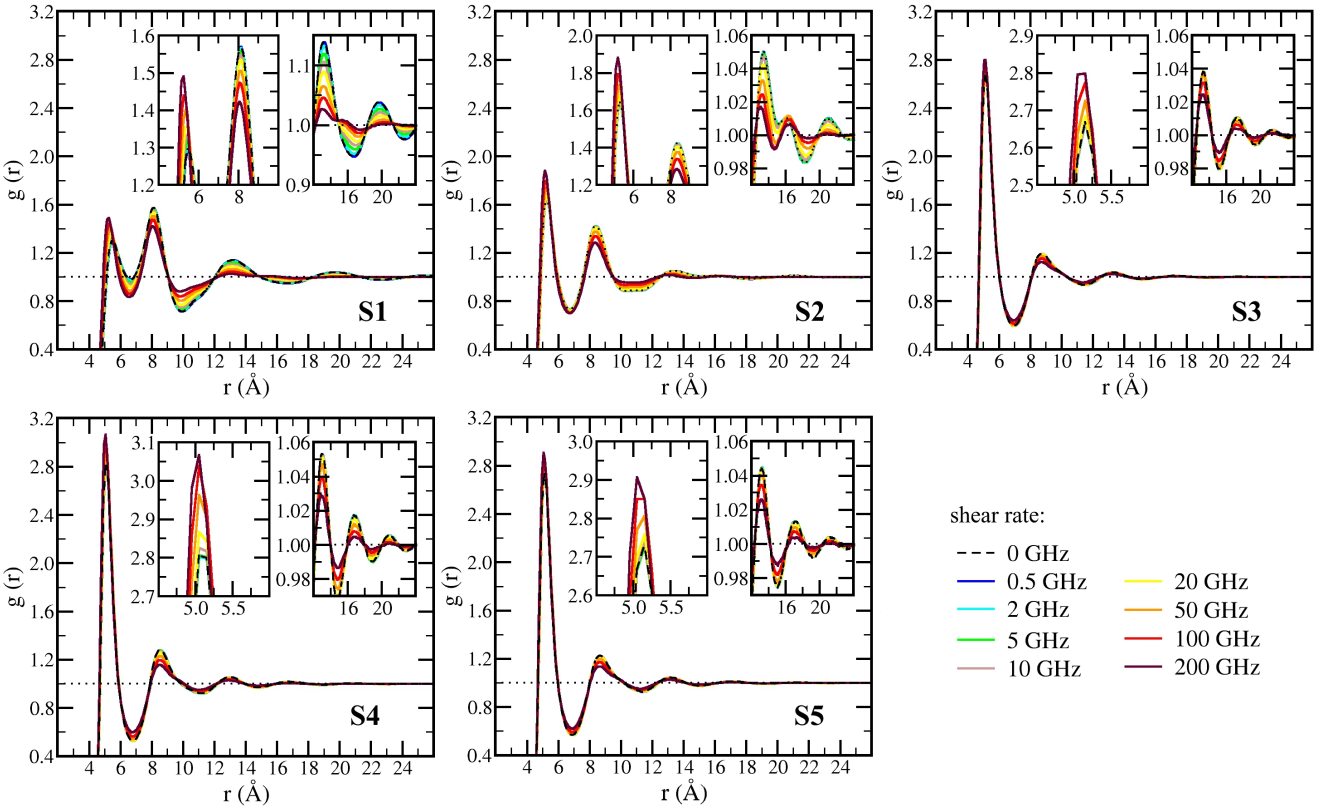

Figure S3 – Anion – anion radial distribution function at several shear rates (different colors) for the five model systems with insets highlighting the first maximum and the long range structure.

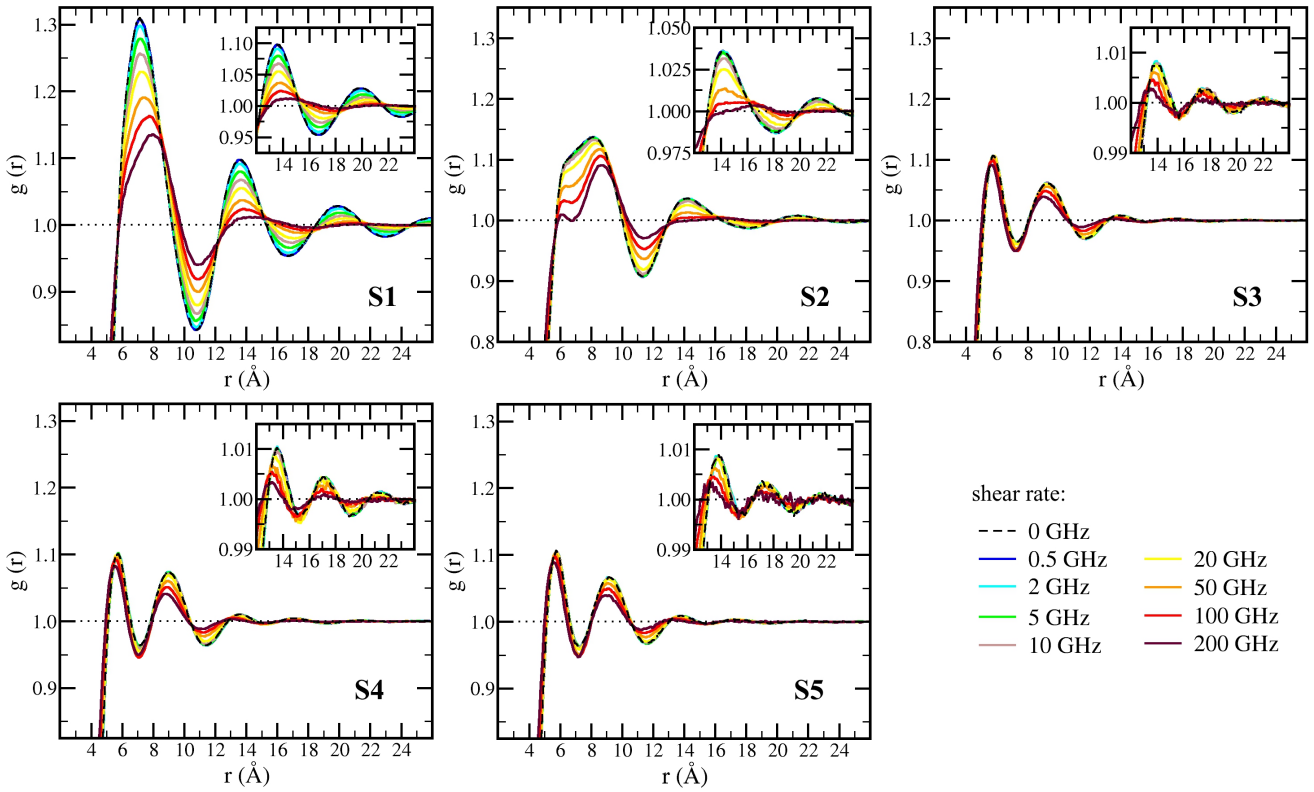

Figure S4 – Cation – cation radial distribution function at several shear rates (different colors) for the five model systems with insets highlighting the first maximum and the long range structure.