Supplementary Material

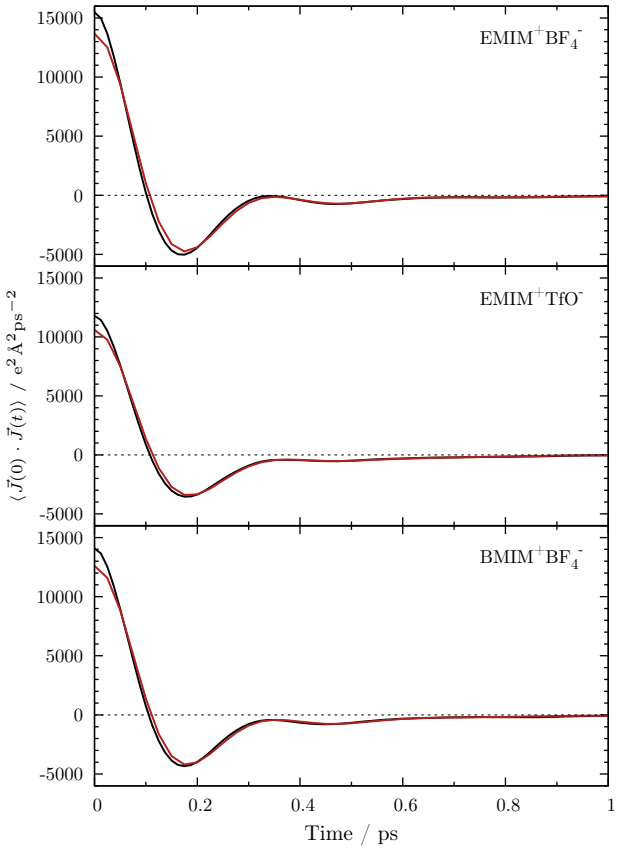

FIG. S1. Comparison of $\langle J(0)J(t)\rangle$ (black) and $\frac{1}{2}\frac{d^2}{dt^2}\langle(\Delta M_J(t))^2\rangle$ (red). This relationship has already been shown in previous work.[1,2]

TABLE S1. Table listing all fit parameters used to describe the time correlation functions underlying the dielectric relaxation data of $EMIM^+BF_4^-$.

$\langle \vec{M}_D(0) \cdot \vec{M}_D(t) \rangle$	A_k	τ_k/ps	ω_k/THz	δ_k
$A_1 e^{-t/\tau_1}$	42.40	2730	–	–
$A_2 e^{-t/\tau_2}$	67.64	421	–	–
$A_3 e^{-t/\tau_3}$	8.951	21.1	–	–
$A_4 e^{-t/\tau_4}$	5.344	2.52	–	–
$A_5 e^{-t/\tau_5}$	5.332	0.400	–	–
$A_6 e^{-t/\tau_6}$	7.323	0.0660	–	–
$\langle \vec{J}(0) \cdot \vec{J}(t) \rangle$	A_k	τ_k/ps	ω_k/THz	δ_k
$A_1 cos(\omega_1 t + \delta_1) \cdot e^{-t/\tau_1}$	14200	0.121	20.159	-0.9027
$A_2 cos(\omega_2 t + \delta_2) \cdot e^{-t/\tau_2}$	406.6	0.0957	34.892	-1.890
$A_3 e^{-t/\tau_3}$	-5358	0.153	–	–
$A_4 e^{-t/\tau_4}$	-816.7	0.311	–	–
$A_5 e^{-t/\tau_5}$	11330	0.0546	–	–
$A_6 e^{-t/\tau_6}$	-139.7	0.904	–	–
$A_7 e^{-t/\tau_7}$	-8.866	4.47	–	–
$A_8 e^{-t/\tau_8}$	-0.2366	40.6	–	–
$\langle \vec{M}_D(0) \cdot \vec{J}(t) \rangle$	A_k	τ_k/ps	ω_k/THz	δ_k
$A_1 cos(\omega_1 t + \delta_1) \cdot e^{-t/\tau_1}$	359.1	0.0969	19.446	-2.375
$A_2 cos(\omega_2 t + \delta_2) \cdot e^{-t/\tau_2}$	798.1	0.0235	11.331	-1.223
$A_3 e^{-t/\tau_3}$	-1.362	10.1	–	–
$A_4 e^{-t/\tau_4}$	-12.83	0.340	–	–

TABLE S2. Table listing all fit parameters used to describe the time correlation functions underlying the dielectric relaxation data of $EMIM^+TfO^-$.

$\langle \vec{M}_D(0) \cdot \vec{M}_D(t) \rangle$	A_k	τ_k/ps	ω_k/THz	δ_k
$A_1 e^{-t/\tau_1}$	208.6	3750	–	–
$A_2 e^{-t/\tau_2}$	220.7	36.3	–	–
$A_3 e^{-t/\tau_3}$	128.0	0.416	–	–
$A_4 e^{-t/\tau_4}$	263.2	241	–	–
$A_5 e^{-t/\tau_5}$	181.4	5.87	–	–
$\langle \vec{J}(0) \cdot \vec{J}(t) \rangle$	A_k	τ_k/ps	ω_k/THz	δ_k
$A_1 cos(\omega_1 t + \delta_1) \cdot e^{-t/\tau_1}$	7953	0.123	20.066	-0.6724
$A_2 cos(\omega_2 t + \delta_2) \cdot e^{-t/\tau_2}$	8461	0.102	11.452	-0.2029
$A_3 e^{-t/\tau_3}$	-2666	0.260	–	–
$A_4 e^{-t/\tau_4}$	-22.00	2.37	–	–
$A_5 e^{-t/\tau_5}$	-0.6411	23.8	–	–
$\langle \vec{M}_D(0) \cdot \vec{J}(t) \rangle$	A_k	τ_k/ps	ω_k/THz	δ_k
$A_1 cos(\omega_1 t + \delta_1) \cdot e^{-t/\tau_1}$	288.9	0.105	19.191	-2.486
$A_2 cos(\omega_2 t + \delta_2) \cdot e^{-t/\tau_2}$	724.6	0.184	4.0888	1.217
$A_3 cos(\omega_3 t + \delta_3) \cdot e^{-t/\tau_3}$	13.29	2.59	0.24729	-2.533
$A_4 e^{-t/\tau_4}$	-4.008	14.5	–	–

TABLE S3. Table listing all fit parameters used to describe the time correlation functions underlying the dielectric relaxation data of $BMIM^+BF_4^-$.

$\langle \vec{M}_D(0) \cdot \vec{M}_D(t) \rangle$	A_k	τ_k/ps	ω_k/THz	δ_k
$A_1 e^{-t/\tau_1}$	47.52	0.316	–	–
$A_2 e^{-t/\tau_2}$	67.53	179	–	–
$A_3 e^{-t/\tau_3}$	31.53	7.51	–	–
$A_4 e^{-t/\tau_4}$	551.2	5140	–	–
$\langle \vec{J}(0) \cdot \vec{J}(t) \rangle$	A_k	τ_k/ps	ω_k/THz	δ_k
$A_1 cos(\omega_1 t + \delta_1) \cdot e^{-t/\tau_1}$	12950	0.115	20.419	-1.015
$A_2 cos(\omega_2 t + \delta_2) \cdot e^{-t/\tau_2}$	99.08	0.119	38.372	-1.937
$A_3 e^{-t/\tau_3}$	-5175	0.213	–	–
$A_4 e^{-t/\tau_4}$	-61.76	1.35	–	–
$A_5 e^{-t/\tau_5}$	11040	0.0563	–	–
$A_6 e^{-t/\tau_6}$	-1.439	9.58	–	–
$A_7 e^{-t/\tau_7}$	-0.3051	41.2	–	–
$A_8 e^{-t/\tau_8}$	0.3995	30.8	–	–
$\langle \vec{M}_D(0) \cdot \vec{J}(t) \rangle$	A_k	τ_k/ps	ω_k/THz	δ_k
$A_1 cos(\omega_1 t + \delta_1) \cdot e^{-t/\tau_1}$	236.9	0.120	19.494	-2.189
$A_2 cos(\omega_2 t + \delta_2) \cdot e^{-t/\tau_2}$	919.9	0.146	1.9590	1.420

REFERENCES

[1] C. Schröder, J. Chem. Phys. **135**, 024502 (2011).

[2] C. Schröder and O. Steinhauser, J. Chem. Phys. **131**, 114504 (2009).