

SUPPORTING INFORMATION

An Inelastic Neutron Scattering, Raman, Far-Infrared, and Molecular Dynamics Study of the Intermolecular Dynamics of Two Ionic Liquids

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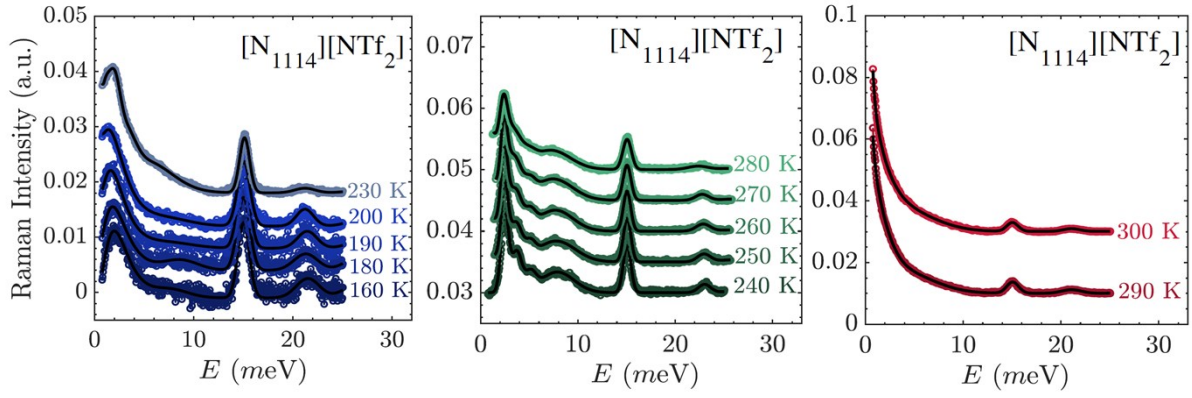


Figure S1. Low-frequency Raman spectra of $[N_{1114}][NTf_2]$ at different temperatures in the glassy (left), crystalline (middle), and liquid (right) phases.

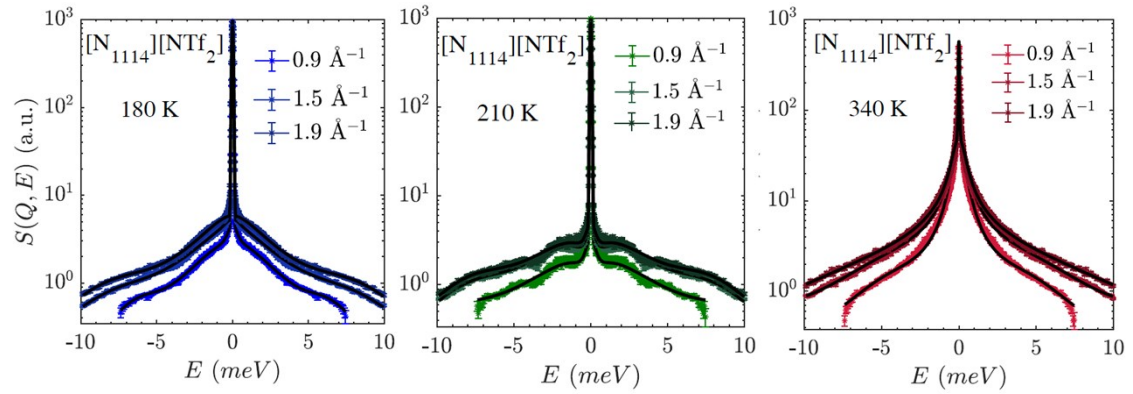


Figure S2. INS spectra of $[N_{1114}][NTf_2]$ at glassy (180 K, left), crystalline (210 K, middle), and liquid (340 K, right) phases for different wavevectors.

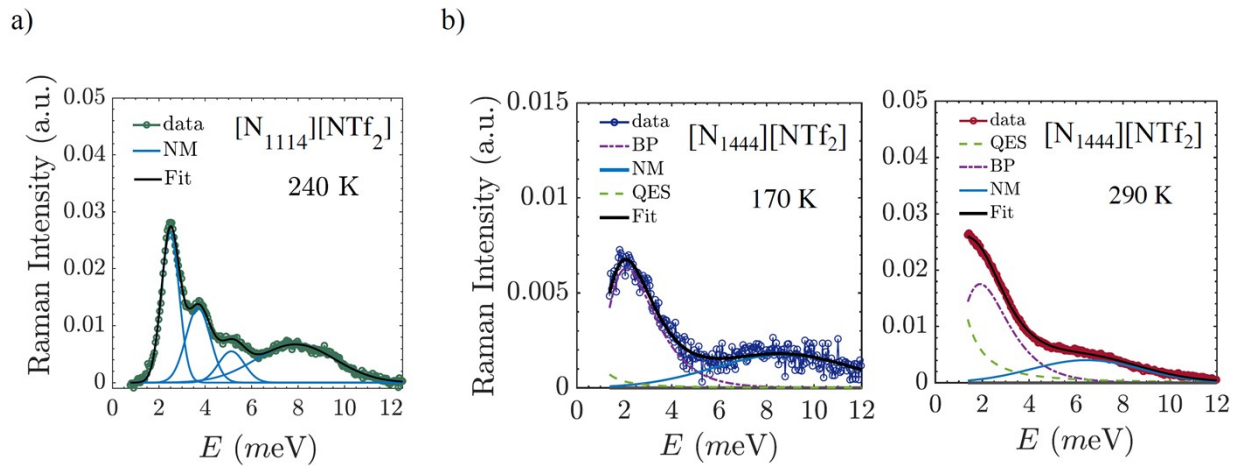


Figure S3. Fits of the Raman spectra of $[\text{N}_{1114}][\text{NTf}_2]$ and $[\text{N}_{1444}][\text{NTf}_2]$: experimental data (symbols), total best fit (black lines), and the components QES (light green dashed line), boson peak (purple dashed line), and additional vibrational bands (light blue line).

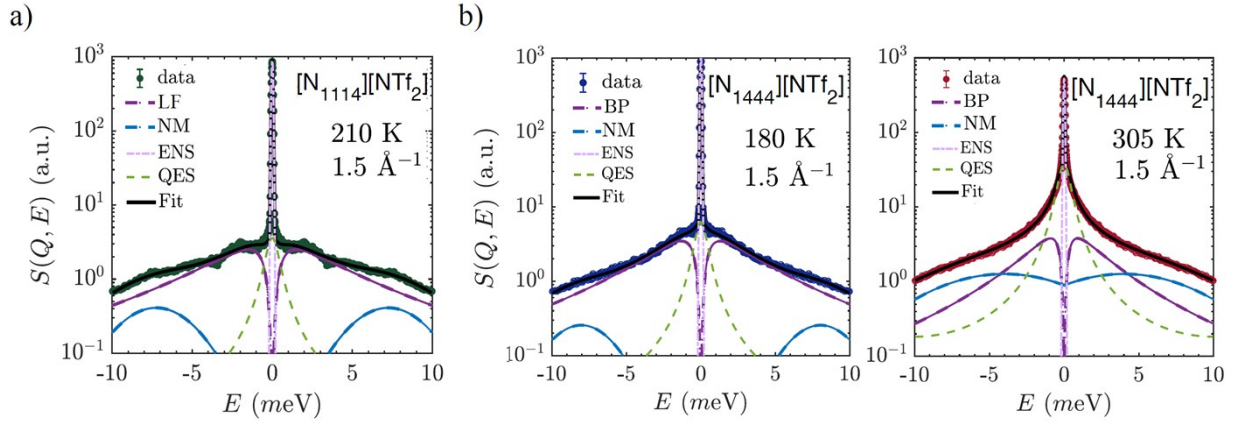


Figure S4. Fits of the INS spectra of $[\text{N}_{1114}][\text{NTf}_2]$ and $[\text{N}_{1444}][\text{NTf}_2]$: experimental data (symbols), total best fit (black lines), and the components QES (light green dashed line), elastic neutron scattering ENS (magenta dashed line), boson peak (purple dashed line), and additional vibrational bands (light blue line).

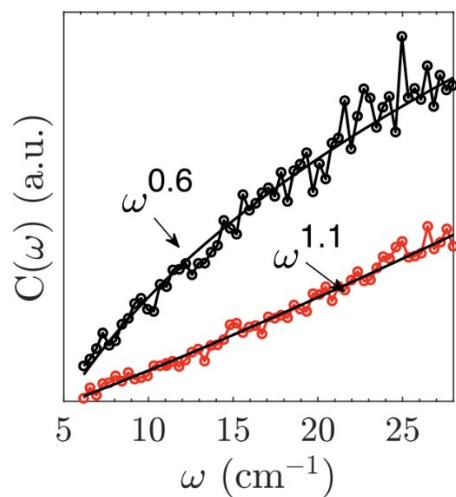


Figure S5. The light-vibration coupling, $C(\omega)$, obtained from the comparison of INS and Raman spectra of $[\text{N}_{1114}][\text{NTf}_2]$ (black symbols) and $[\text{N}_{1444}][\text{NTf}_2]$ (red symbols). The black solid curves are the fitted curves.

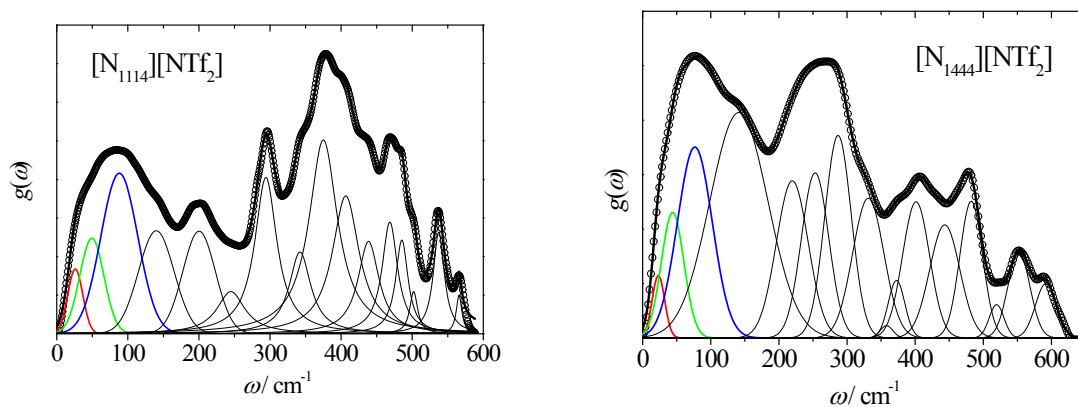


Figure S6. Vibrational density of states, $g(\omega)$, calculated by MD simulations of $[\text{N}_{1114}][\text{NTf}_2]$ (left) and $[\text{N}_{1444}][\text{NTf}_2]$ (right). The bold black line is the total fit on the MD data shown as circles. The components of the fits with peak frequency below 100 cm^{-1} are shown by red, green, and blue lines, and those beyond 100 cm^{-1} are shown by thin black lines.

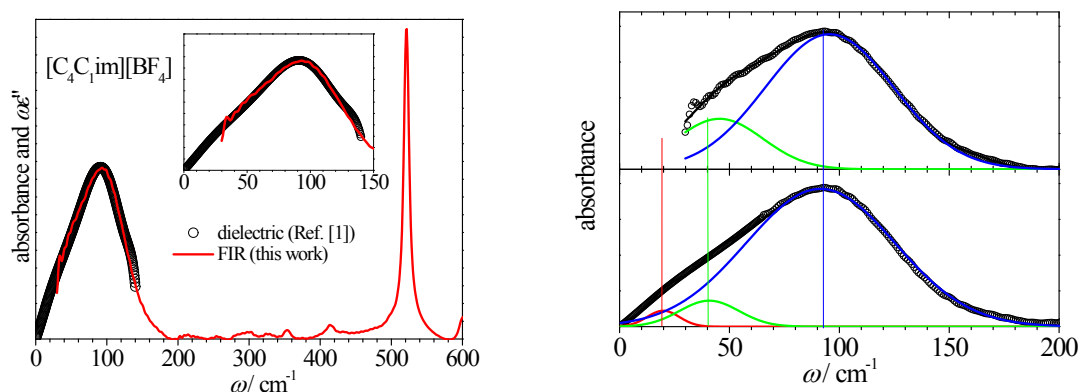


Figure S7. Left panel: FIR spectrum (red line in the 30–600 cm^{-1} range; this work) and the imaginary part of the complex dielectric spectrum, $\omega \cdot \epsilon''(\omega)$ (symbols in the 0–140 cm^{-1} range, Ref. [1]) of $[\text{C}_4\text{C}_1\text{im}][\text{BF}_4]$. The inset highlights the low-frequency range. Right panel: The bold black line is the total fit on the experimental FIR and dielectric data shown as circles. The top panel is the fit using only the FIR spectrum, and the bottom panel is the fit using the combined FIR and dielectric data. The components of the fits with peak frequency below 100 cm^{-1} are shown by red, green, and blue lines.

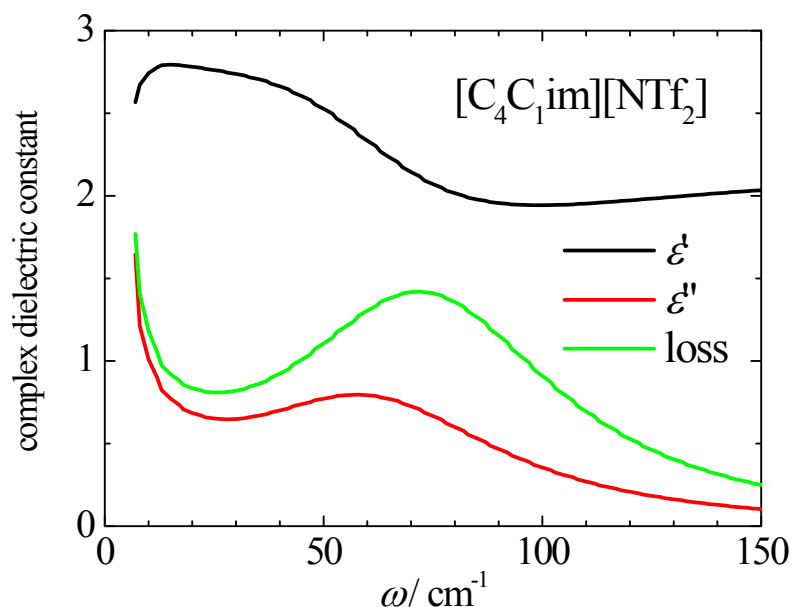


Figure S8. The real (ϵ' , black line) and the imaginary (ϵ'' , red line) parts of the complex dielectric spectra of $[\text{C}_4\text{C}_1\text{im}][\text{NTf}_2]$ given in Ref. [2]. The green line is the resulting

dielectric loss spectrum calculated as $\varepsilon''^2/[\varepsilon'^2+\varepsilon''^2]$. The imaginary and the loss components account for the transverse and the longitudinal dielectric susceptibilities, respectively.

REFERENCES

- (1) K. Yamamoto, M. Tani, M. Hangyo, *J. Phys. Chem. B*, 2007, **111**, 4854-4859.
- (2) S. Mou, A. Rubano, D. Paparo, *J. Phys. Chem. B*, 2017, 121, 7351-7358.