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An inelastic neutron scattering, Raman, far-infrared, and molecular dynamics study of the intermolecular dynamics of two ionic liquids†

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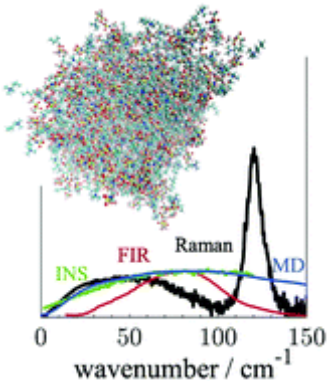
Abstract

The intermolecular dynamics in the THz frequency range of the ionic liquids *n*-butyl-trimethylammonium bis(trifluoromethanesulfonyl)imide, $[N_{1114}][NTf_2]$, and methyl-tributylammonium bis(trifluoromethanesulfonyl)imide, $[N_{1444}][NTf_2]$, were investigated by a combined usage of inelastic neutron scattering (INS), Raman, and far-infrared (FIR) spectroscopies and the power spectrum calculated by molecular dynamics (MD) simulations. The collective dynamics of the simulated systems is also discussed by the calculation of time correlation functions of charge and mass currents that are projected onto acoustic- and optic-like motions. The INS and Raman measurements have been performed as a function of temperature in the glassy, crystalline, and liquid phases. The excess in the vibrational density of states over the expectation of the Debye theory, the so-called boson peak, is found in the INS and Raman spectra as a peak at ~2 meV (~16 cm^{-1}) and also in the direct measurement of heat capacity at very low temperatures (4–20 K). This low-frequency vibration is incorporated into the curve fits of Raman, FIR, and MD data at room temperature. Fits of spectra from these different sources in the range below 100 cm^{-1} are consistently achieved with three components at *ca.* 25, 50, and 80 cm^{-1}, but with distinct relative intensities among the different techniques. It is

proposed as the collective nature of the lowest-frequency component and the anion–cation intermolecular vibration nature of the highest-frequency component. The MD results indicate that there is no clear distinction between acoustic and optic vibrations in the spectral range investigated in this work for the ionic liquids $[N_{1114}][NTf_2]$ and $[N_{1444}][NTf_2]$. The analysis carried out here agrees in part, but not entirely, with other propositions in the literature, mainly from optical Kerr effect (OKE) and FIR spectroscopies, concerning the intermolecular dynamics of ionic liquids.



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