Supporting Information

Sum Frequency Generation Spectroscopy of Imidazolium-based Ionic Liquids with Cyano-functionalized Anions at the Solid Salt-Liquid Interface Chariz Y. Peñalber, Gary A. Baker,† and Steven Baldelli* Department of Chemistry, University of Houston, Houston, Texas 77204-5003 †Department of Chemistry, University of Missouri-Columbia, Columbia, Missouri, 65211-7600 sbaldelli@uh.edu*



Fig. 1S. SFG spectra of IL–NaCl{100} interface: A) near the C–H stretching region in *pss*; and B) near the C–N stretching region (re-scaled for clarity) in *ssp* and *ppp* polarizations.

[BMIM][DCA]- BaF₂(111) [BMIM][TCM]– BaF₂(111) $[BMIM][SCN]-BaF_2(111)$ A) C-H Region 0.15· 0.15 -1.2 0.12 Normalized SFG Intensity 0.12 0.9 -o-ssp ssp ■ ssp 0.09 0.09 0.6 0.06 0.06 0.3 0.03 0.03 1. Paker 2 0.00 0.00 0.0 3300 3150 2700 2850 3000 3150 3300 2700 2850 3000 2700 2850 3150 3300 3000 IR Wavenumber, cm⁻¹ IR Wavenumber, cm⁻¹ IR Wavenumber, cm⁻¹ 0.15 1.2 0.15 0.12 0.12 0.9 Normalized SFG Intensity -o-ppp ppp 0.09 --- ppp 0.09 0.6 0.06 0.06 ીંગુકુલ જીવુ 0.3 0.03 0.03 0.00 0.00 0.0 2700 2850 3000 3150 3300 2700 2850 3000 3150 3300 2700 2850 3000 3150 IR Wavenumber, cm⁻¹ IR Wavenumber, cm⁻¹ IR Wavenumber, cm⁻¹ B) C-N Region 2.4 2.4 2.4



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Fig. 2S. SFG spectra of IL-BaF₂(111) interface: A) near the C-H stretching region in ssp and ppp ([BMIM][TCM] re-scaled due to high background from colored sample); and B) near the C-N stretching region in pss polarization.



Fig. 3S. SFG spectra of [EMIM[TCB]–BaF₂(111) interface near the C–H stretching region in *pss*.



Fig. 4S. SFG spectra of [BMIM][SCN]–BaF₂(111) interface near the C–N stretching region at all polarization combinations.



Fig. 5S. SFG spectra of IL-NaCl(100) interface zoomed in from 2750 to 3000 cm⁻¹ region for clarity, showing all polarization combinations. Solid lines indicate a fit to the data in (▲) for [BMIM][SCN], (●) for [BMIM][DCA] and (■) for [BMIM][TCM] using equation 3 as a fitting function