## Supporting Information

Sum Frequency Generation Spectroscopy of Imidazolium-based Ionic Liquids with Cyano-functionalized Anions at the Solid Salt-Liquid Interface

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[BMIM][SCN]-NaCl $\{100\}$
A) $\mathrm{C}-\mathrm{H}$ Region
[BMIM][DCA]-NaCl $\{100\}$
[BMIM][TCM]-NaCl $\{100\}$

B) $\mathrm{C}-\mathrm{N}$ Region


Fig. 1S. SFG spectra of $\mathrm{IL}-\mathrm{NaCl}\{100\}$ interface: A) near the $\mathrm{C}-\mathrm{H}$ stretching region in $p s s$; and B) near the $\mathrm{C}-\mathrm{N}$ stretching region (re-scaled for clarity) in $s s p$ and $p p p$ polarizations.
A) $\mathrm{C}-\mathrm{H}$ Region

B) C-N Region




Fig. 2S. SFG spectra of $\mathrm{IL}^{-} \mathrm{BaF}_{2}(111)$ interface: A) near the $\mathrm{C}-\mathrm{H}$ stretching region in $s s p$ and $p p p$ ([BMIM][TCM] re-scaled due to high background from colored sample); and B ) near the $\mathrm{C}-\mathrm{N}$ stretching region in pss polarization.
[EMIM][TCB]- $\mathrm{BaF}_{2}(111)$


Fig. 3S. SFG spectra of $\left[E M I M[T C B]-\mathrm{BaF}_{2}(111)\right.$ interface near the $\mathrm{C}-\mathrm{H}$ stretching region in $p s s$.


Fig. 4S. SFG spectra of $[\mathrm{BMIM}][\mathrm{SCN}]-\mathrm{BaF}_{2}(111)$ interface near the $\mathrm{C}-\mathrm{N}$ stretching region at all polarization combinations.


Fig. 5S. SFG spectra of $\mathrm{IL}-\mathrm{NaCl}(100)$ interface zoomed in from 2750 to $3000 \mathrm{~cm}^{-1}$ region for clarity, showing all polarization combinations. Solid lines indicate a fit to the data in $(\mathbf{\Delta})$ for $[\mathrm{BMIM}][\mathrm{SCN}],(\bullet)$ for $[\mathrm{BMIM}][\mathrm{DCA}]$ and $(■)$ for $[\mathrm{BMIM}][\mathrm{TCM}]$ using equation 3 as a fitting function

