*Electronic Supplementary Information:*

# The structure of a lanthanide complex at an extractant/water interface studied using heterodyne-detected vibrational sum frequency generation

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**HDEHP concentration dependence of the Im$\chi^{(2)}$ spectrum of the HDEHP/water interface in the presence of $Eu^{3+}$**

Figure S1 shows the Im$\chi^{(2)}$ spectra of the HDEHP/water interfaces in the presence of $Eu^{3+}$ (10 mM). The three samples have different HDEHP concentration. All spectra exhibit negative water OH bands identical within experimental error. This result indicates that HDEHP cannot be bonded to $Eu^{3+}$ from the water phase side even at the saturated concentration of HDEHP, which is different from the case of DBP.

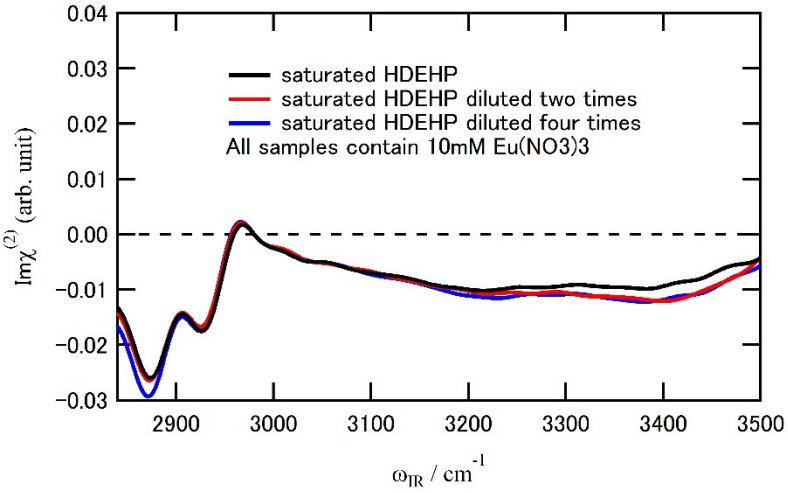


**Figure S1.** Im$\chi^{(2)}$ spectra of the HDEHP/aqueous interfaces of $Eu(NO_3)_3$ solutions. The spectral features do not depend on the concentration of HDEHP within experimental error. 10 mM $Eu(NO_3)_3$ is dissolved in all solutions.