

Supporting Information:
**Effects of Multivalent Hexacyanoferrates and
Their Ion Pairs on Water Molecule Dynamics
Measured with Terahertz Spectroscopy**

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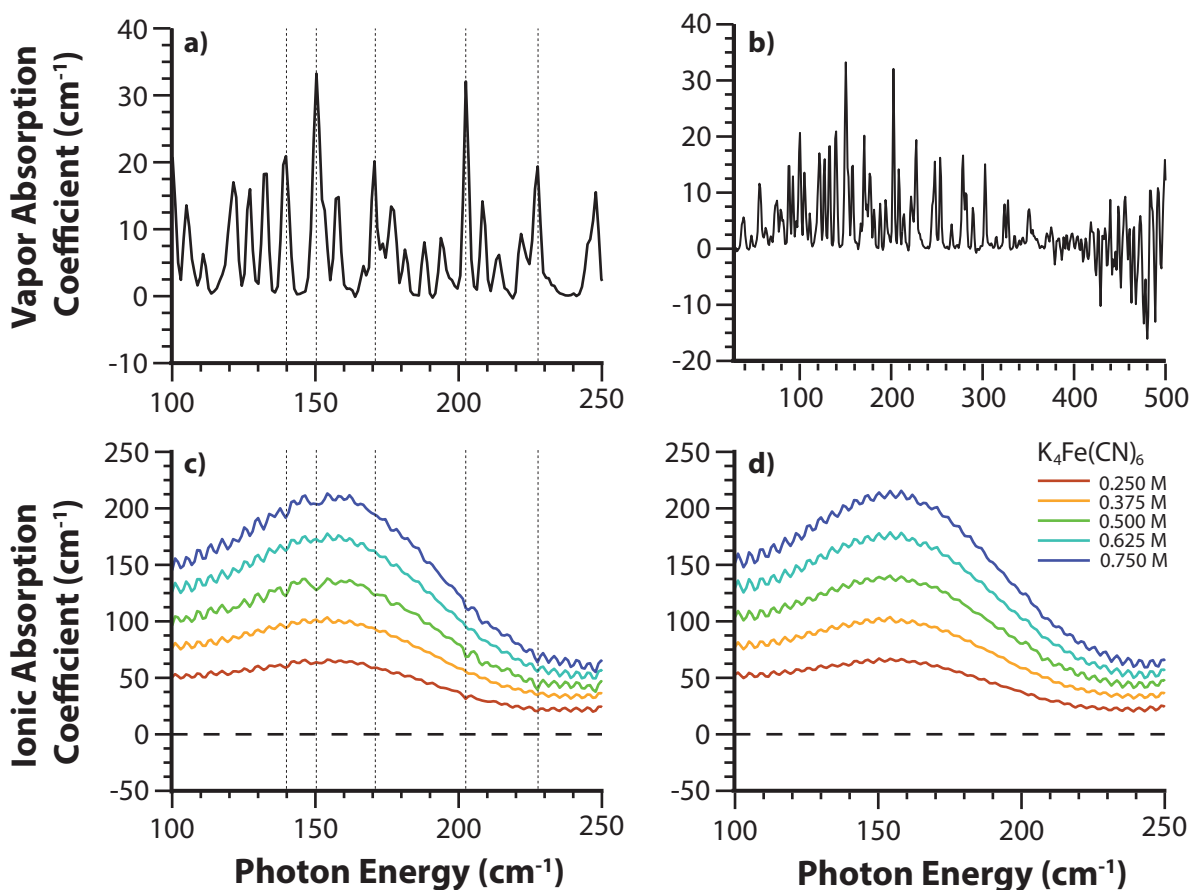


Figure 1: [Figure S1] Illustration of the procedure for subtraction of (a,b) the vapor absorption coefficient from (b) the effective ion absorption coefficient to produce (c) the vapor-corrected absorption coefficient for $\text{K}_4\text{Fe}(\text{CN})_6$. Slight fluctuations in *c*, such as those at ~ 150.4 and 202.5 cm^{-1} , are removed with this scaled vapor removal, which smooths the lineshape for an improved spectral fitting. Vertical dotted lines in *a* and *c* show regions of high intensity in the vapor spectrum correspond to regions with the highest fluctuation in the non-corrected absorption coefficient spectrum.

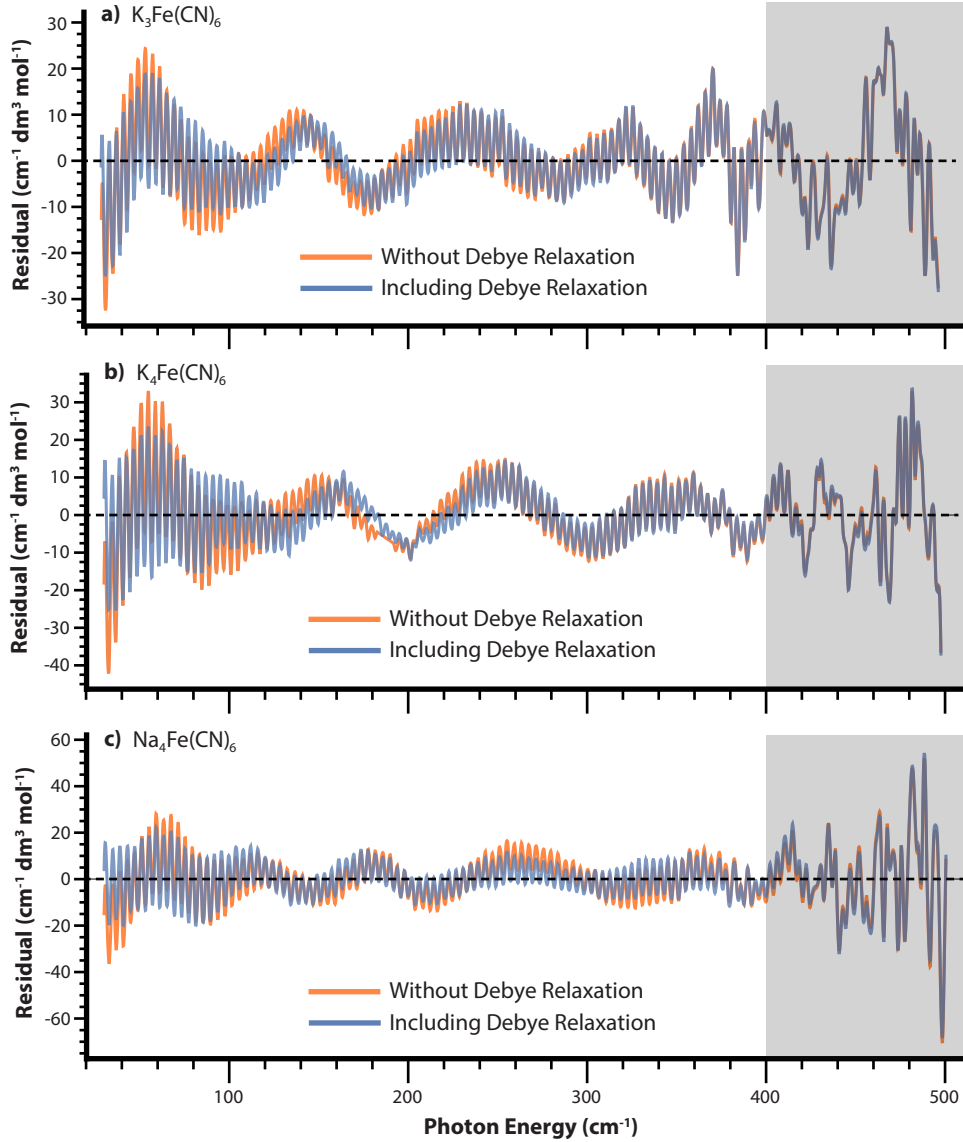


Figure 2: [Figure S2] Illustration of the residuals resulting from damped harmonic fits of the extinction coefficients without (orange) and with (blue) using the low-frequency Debye relaxation mode, $\varepsilon_{Debye}(\tilde{\nu})$. This component was first manually added until the low-frequency region matched the largest residual below 400 cm^{-1} (where laser saturation begins to result in variance) for (a) $\text{K}_3\text{Fe}(\text{CN})_6$, which resulted in an increase of ~ 6 water molecules for $n_{Hydration}$. Addition of $\varepsilon_{Debye}(\tilde{\nu})$ in (b) $\text{K}_4\text{Fe}(\text{CN})_6$ was increased until the residual matched that for a in this region. Only minor addition of $\varepsilon_{Debye}(\tilde{\nu})$ was required for the same residual in the low-frequency region of (c) $\text{Na}_4\text{Fe}(\text{CN})_6$ and further addition did not largely benefit the residual. However, the amplitude of the ion-pairing peak (a_2) is sensitive to the chosen amplitude for $\varepsilon_{Debye}(\tilde{\nu})$, and was therefore adjusted until a_2 was $\sim 85\%$ of the same peak in $\text{K}_4\text{Fe}(\text{CN})_6$, which is consistent with this region of $\varepsilon_{ion}^{eff}(\tilde{\nu})$ for $\text{Na}_4\text{Fe}(\text{CN})_6$ versus $\text{K}_4\text{Fe}(\text{CN})_6$.