

Supporting Information

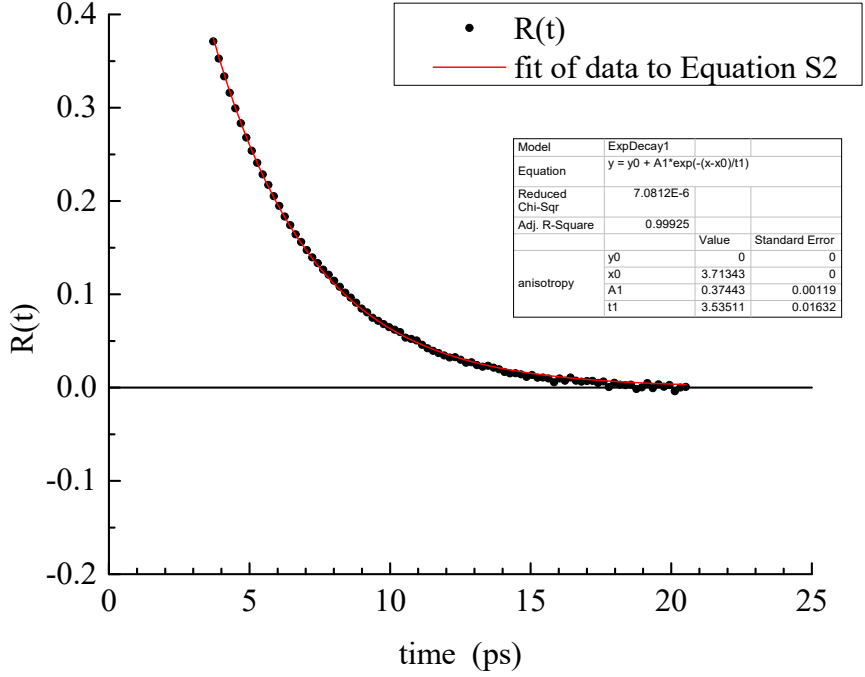
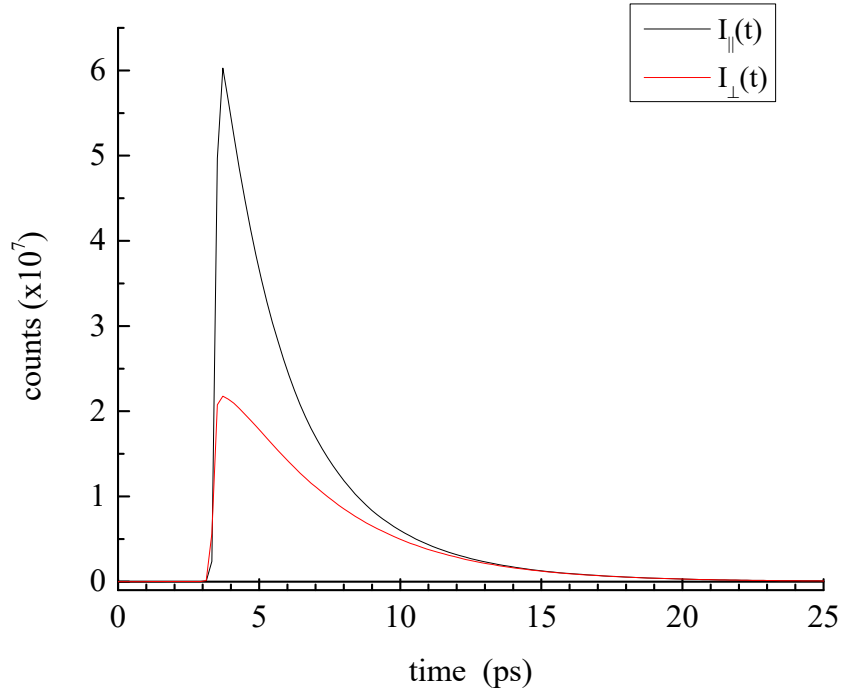
The Effect of Dilution on Induced Free Charge Density Gradients in Room Temperature Ionic Liquids

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Example of experimental data for CV⁺ in BMIM⁺BF₄⁻ (X=0.603) in ACN (X=0.397). Top panel show raw experimental I_{||}(t) and I_⊥(t) data. Bottom panel shows R(t) calculated using Eq. S1 and the fit of the function S2 to the experimental data.

$$R(t) = \frac{I_{\parallel}(t) - I_{\perp}(t)}{I_{\parallel}(t) + 2I_{\perp}(t)} \quad [\text{S1}]$$



$$R(t) = R(0) \exp(-t/\tau_{OR})$$

[S2]

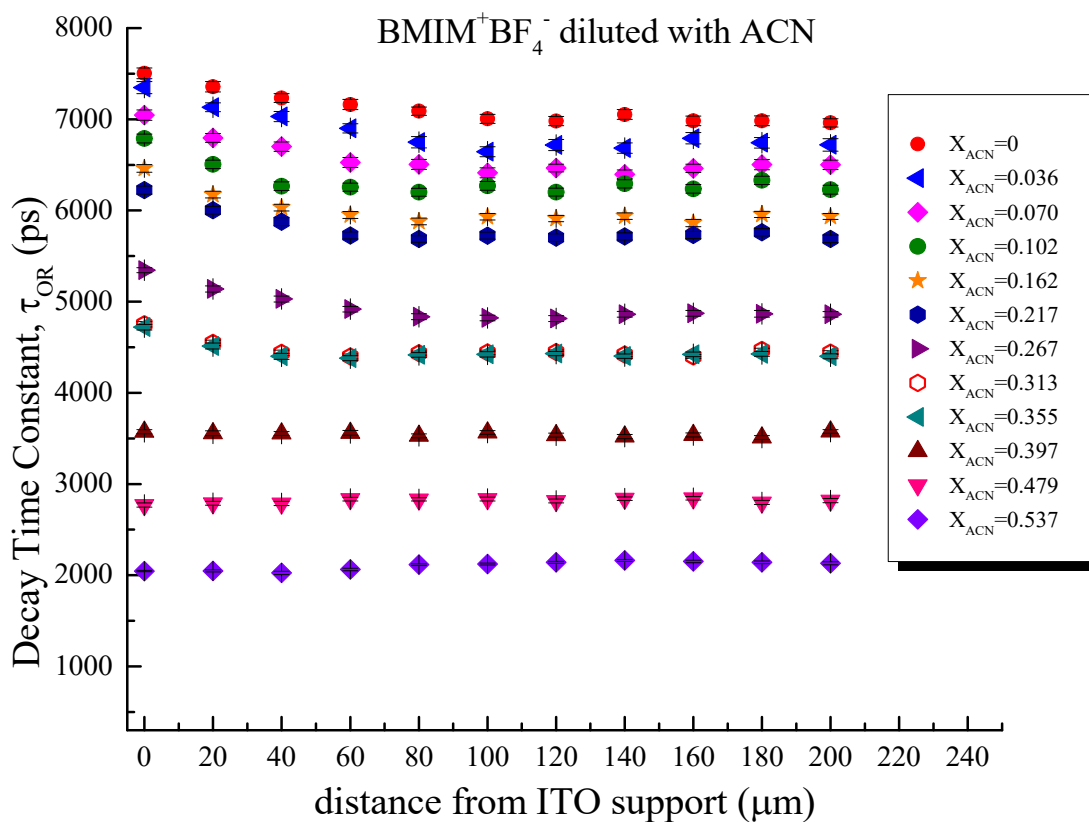


Figure S1. Anisotropy decay time constants of CV⁺ in BMIM⁺BF₄⁻ as a function of distance from the ITO support and as a function of dilution with ACN as indicated in the legend. Each time constant is the result of the average of 65,536 (256 x 256 array) I_{||}(t) and I_⊥(t) time-resolved data sets. The resulting data are processed according to Eq. 1 and the resulting R(t) function is fitted to a single exponential decay function. The error bars are $\pm 1\sigma$ uncertainties for fits of R(t) to a single exponential decay function. Replicate measurements were performed at least three times.

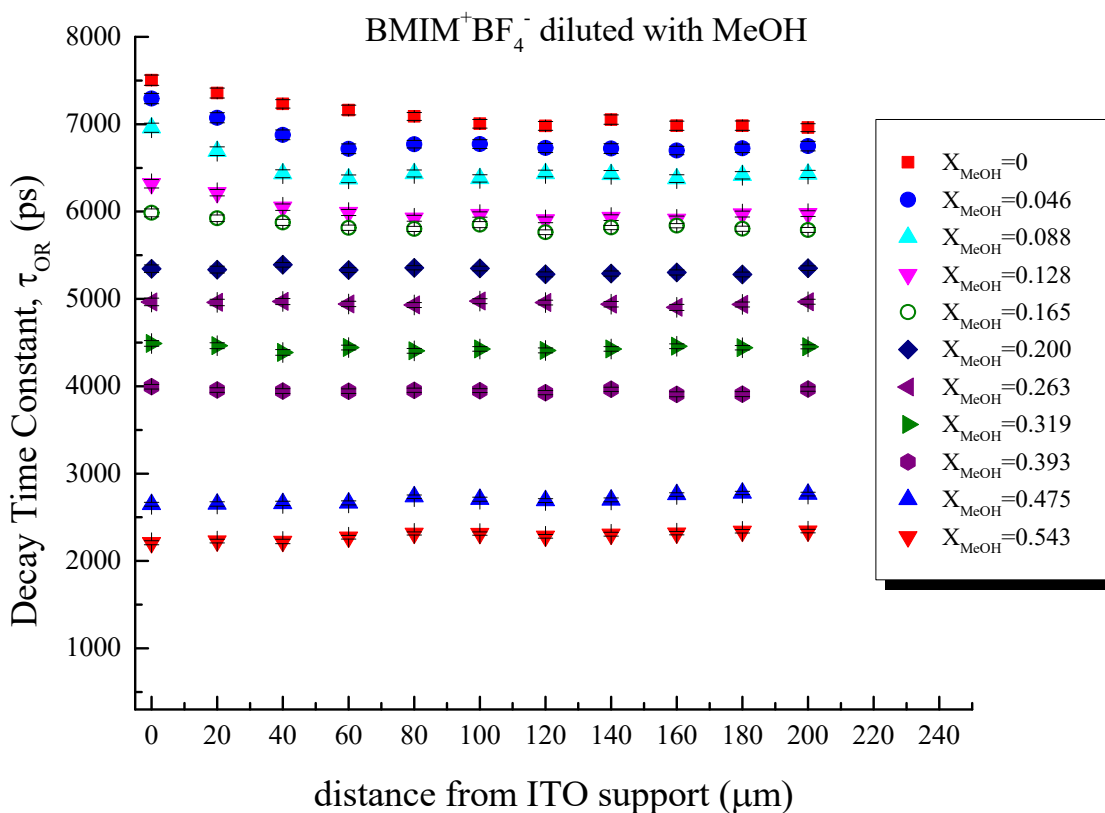


Figure S2. Anisotropy decay time constants of CV⁺ in BMIM⁺BF₄⁻ as a function of distance from the ITO support and as a function of dilution with MeOH as indicated in the legend. Each time constant is the result of the average of 65,536 (256 x 256 array) I_{||}(t) and I_⊥(t) time-resolved data sets. The resulting data are processed according to Eq. 1 and the resulting R(t) function is fitted to a single exponential decay function. The error bars are $\pm 1\sigma$ uncertainties for fits of R(t) to a single exponential decay function. Replicate measurements were performed at least three times.

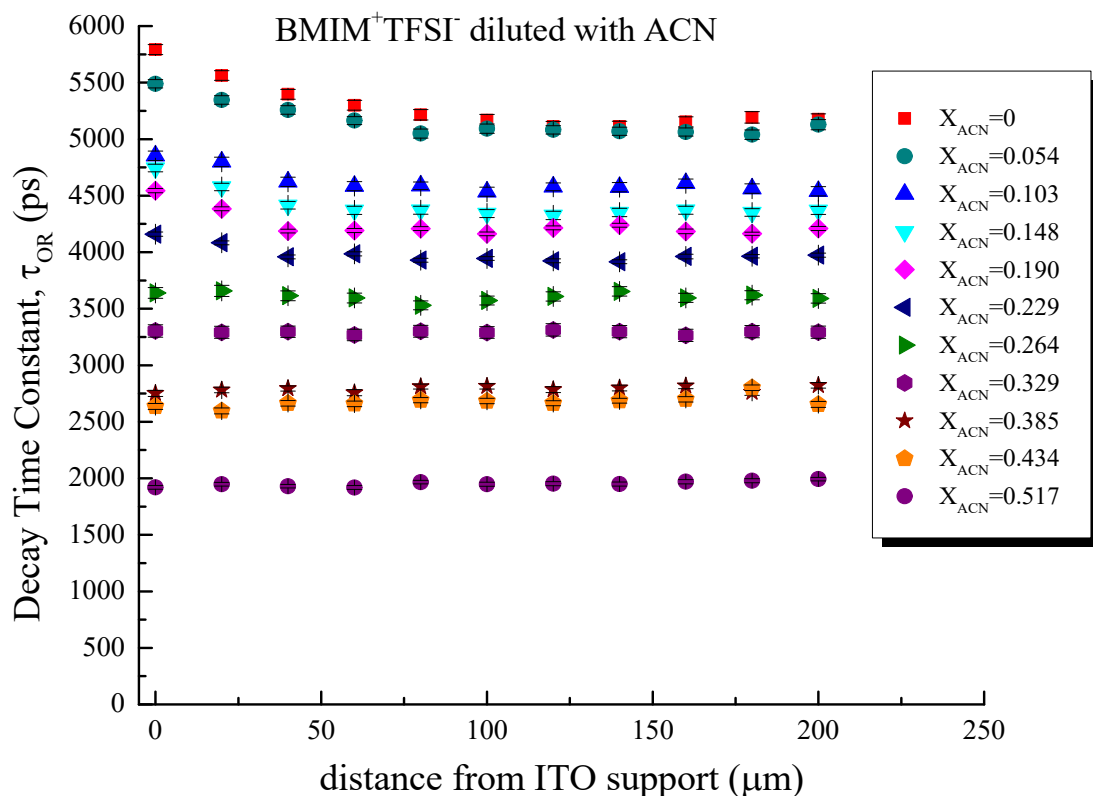


Figure S3. Anisotropy decay time constants of CV⁺ in BMIM⁺TFSI⁻ as a function of distance from the ITO support and as a function of dilution with ACN as indicated in the legend. Each time constant is the result of the average of 65,536 (256 x 256 array) I_{||}(t) and I_⊥(t) time-resolved data sets. The resulting data are processed according to Eq. 1 and the resulting R(t) function is fitted to a single exponential decay function. The error bars are $\pm 1\sigma$ uncertainties for fits of R(t) to a single exponential decay function. Replicate measurements were performed at least three times.

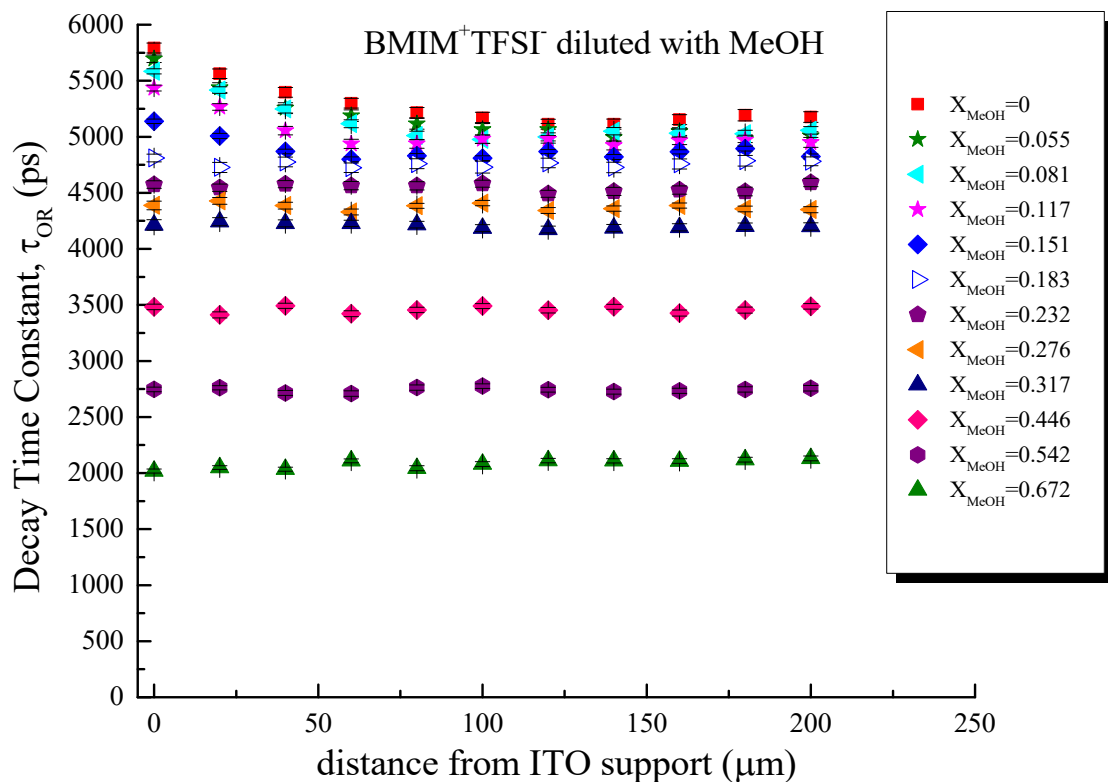


Figure S4. Anisotropy decay time constants of CV⁺ in BMIM⁺TFSI⁻ as a function of distance from the ITO support and as a function of dilution with MeOH as indicated in the legend. Each time constant is the result of the average of 65,536 (256 x 256 array) I_{||}(t) and I_⊥(t) time-resolved data sets. The resulting data are processed according to Eq. 1 and the resulting R(t) function is fitted to a single exponential decay function. The error bars are ±1σ uncertainties for fits of R(t) to a single exponential decay function. Replicate measurements were performed at least three times.

HMIM⁺TFSI⁻ diluted with ACN

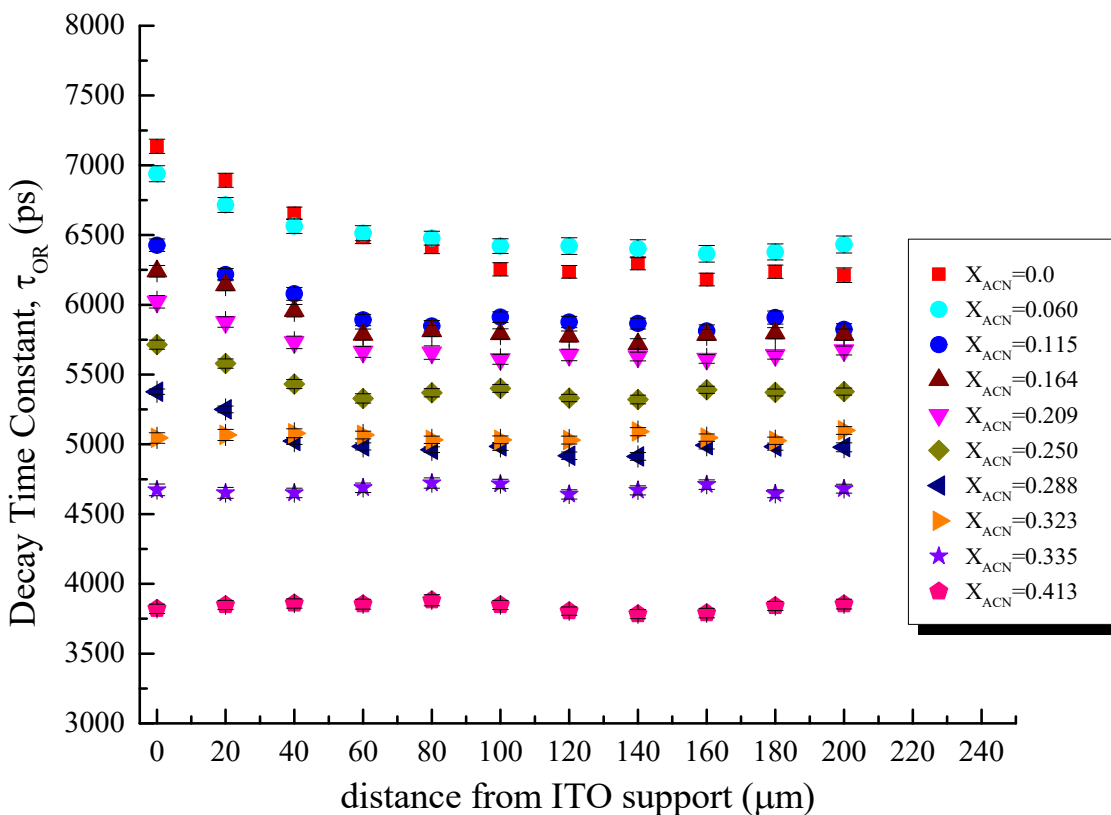


Figure S5. Anisotropy decay time constants of CV⁺ in HMIM⁺TFSI⁻ as a function of distance from the ITO support and as a function of dilution with ACN as indicated in the legend. Each time constant is the result of the average of 65,536 (256 x 256 array) $I_{||}(t)$ and $I_{\perp}(t)$ time-resolved data sets. The resulting data are processed according to Eq. 1 and the resulting $R(t)$ function is fitted to a single exponential decay function. The error bars are $\pm 1\sigma$ uncertainties for fits of $R(t)$ to a single exponential decay function. Replicate measurements were performed at least three times.

HMIM⁺TFSI⁻ diluted with MeOH

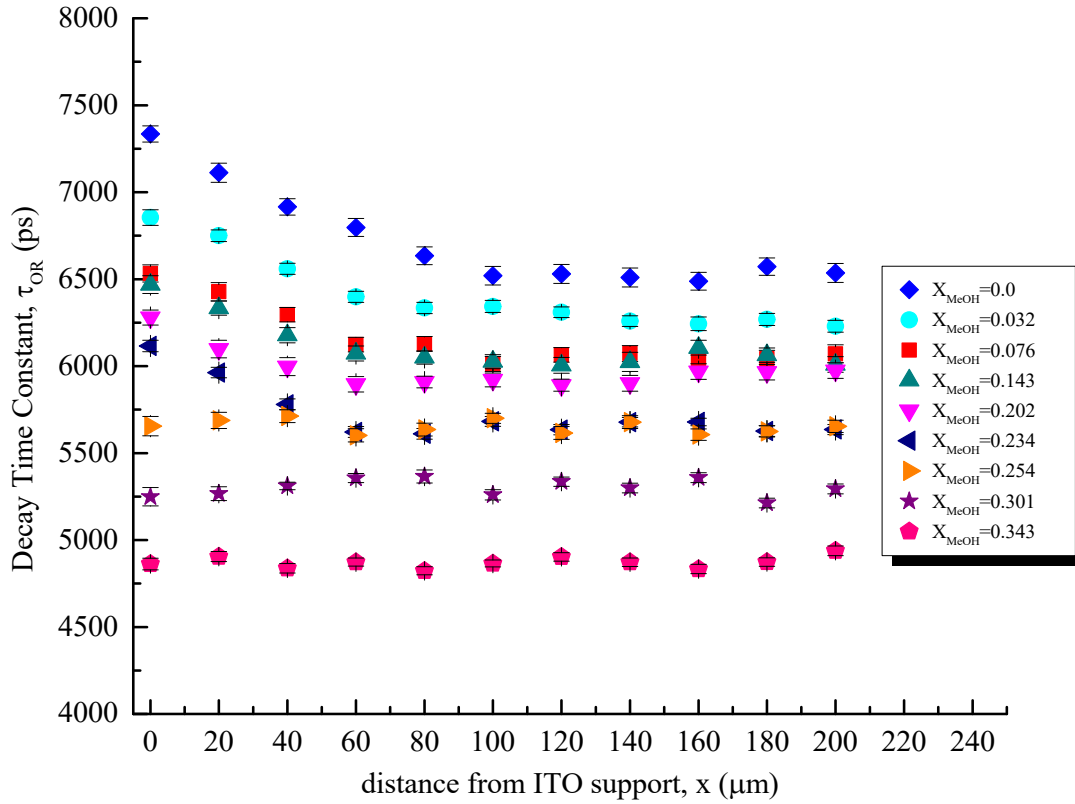


Figure S6. Anisotropy decay time constants of CV^+ in $\text{HMIM}^+\text{TFSI}^-$ as a function of distance from the ITO support and as a function of dilution with MeOH as indicated in the legend. Each time constant is the result of the average of 65,536 (256×256 array) $I_{\parallel}(t)$ and $I_{\perp}(t)$ time-resolved data sets. The resulting data are processed according to Eq. 1 and the resulting $R(t)$ function is fitted to a single exponential decay function. The error bars are $\pm 1\sigma$ uncertainties for fits of $R(t)$ to a single exponential decay function. Replicate measurements were performed at least three times.