

Representative Examples of RE Measurements

We show a representative collection of relaxation data for the four sample pairs 68K1-109[Dy(DOTA)]/68K1-109[Lu(DOTA)], 68K1-109[Dy(DTPA)]/68K1-109[Lu(DOTA)], 131K1-109[Dy(DOTA)]/131K1-109[Lu(DOTA)] and 131K1-109[Dy(DTPA)]/131K1-109[Lu(DOTA)] for temperatures $T=\{40\text{K}, 60\text{K}, 80\text{K}, 100\text{K}\}$. The concentrations of the samples have been estimated with CW EPR measurements during the sample preparation process. All samples were prepared as a mixture of 25 μL glycerol and 25 μL of protein solution in buffer, and had therefore a total sample volume of 50 μL .

Table S1: Sample types and concentrations of nitroxide-labeled (C[NO]) and doubly nitroxide- and Dy^{III}-labeled (C[NO-Dy^{III}]) T4 lysozyme molecules:

Sample	C[NO]	C[NO-Dy ^{III}]	Figure
68K1-109[Lu(DOTA)]	120 μM	-	
68K1-109[Lu(DTPA)]	75 μM	-	
68K1-109[Dy(DOTA)]	130 μM	34 μM	Fig. S1
68K1-109[Dy(DTPA)]	75 μM	14 μM	Fig. S2
131K1-109[Dy(DOTA)]	75 μM	26 μM	Fig. S3
131K1-109[Dy(DTPA)]	75 μM	25 μM	Fig. S4

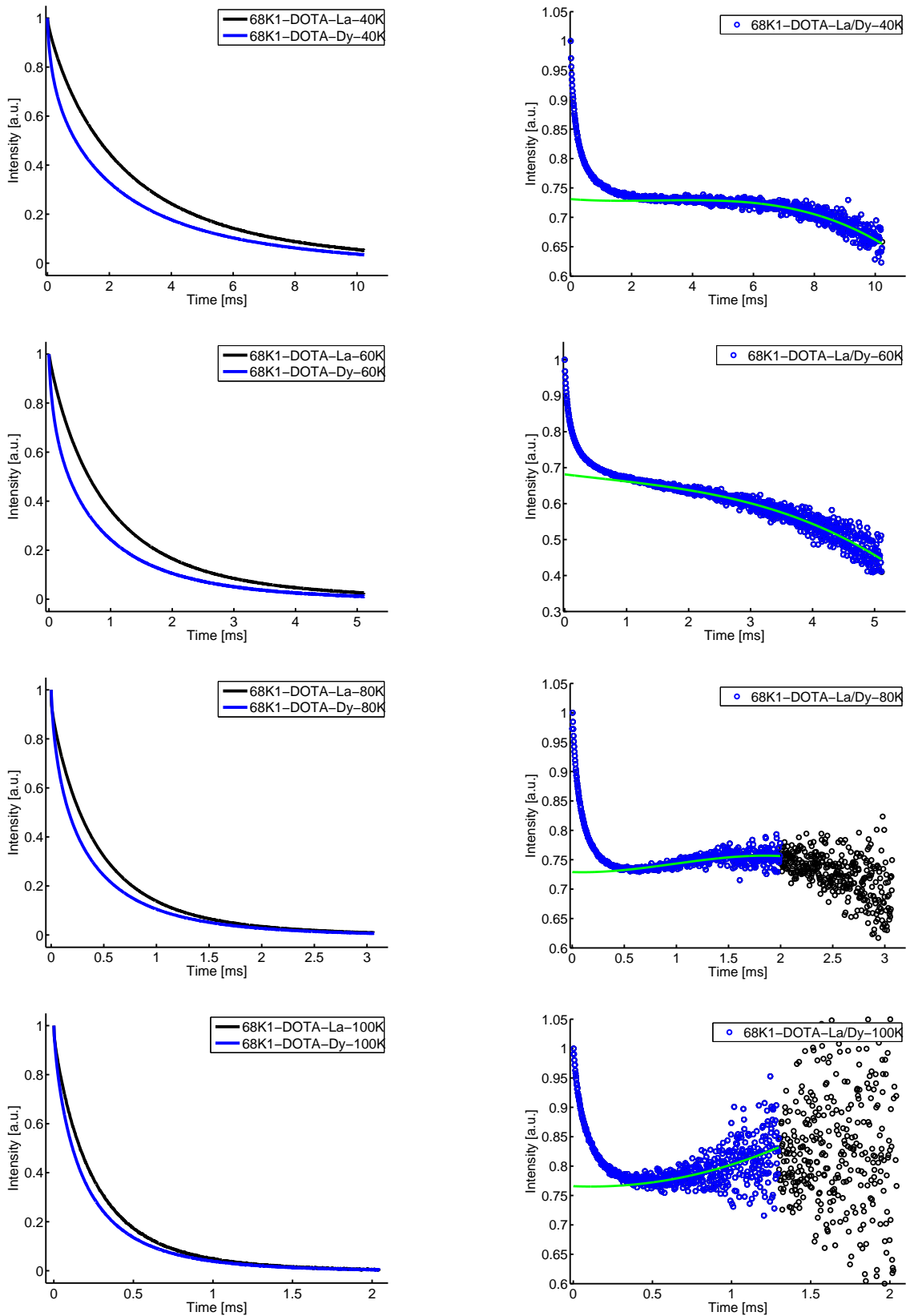


Figure S1: Relaxation data for sample 68K1-109[Dy(DOTA)] for temperatures $T=\{40\text{K}, 60\text{K}, 80\text{K}, 100\text{K}\}$. Left column of figures: Renormalized inversion recovery traces (Dy - blue, Lu - black). Right column of figures: Division of inversion recovery traces Dy/Lu and background fits (green). RE data points cutted out before background fitting are shown in black.

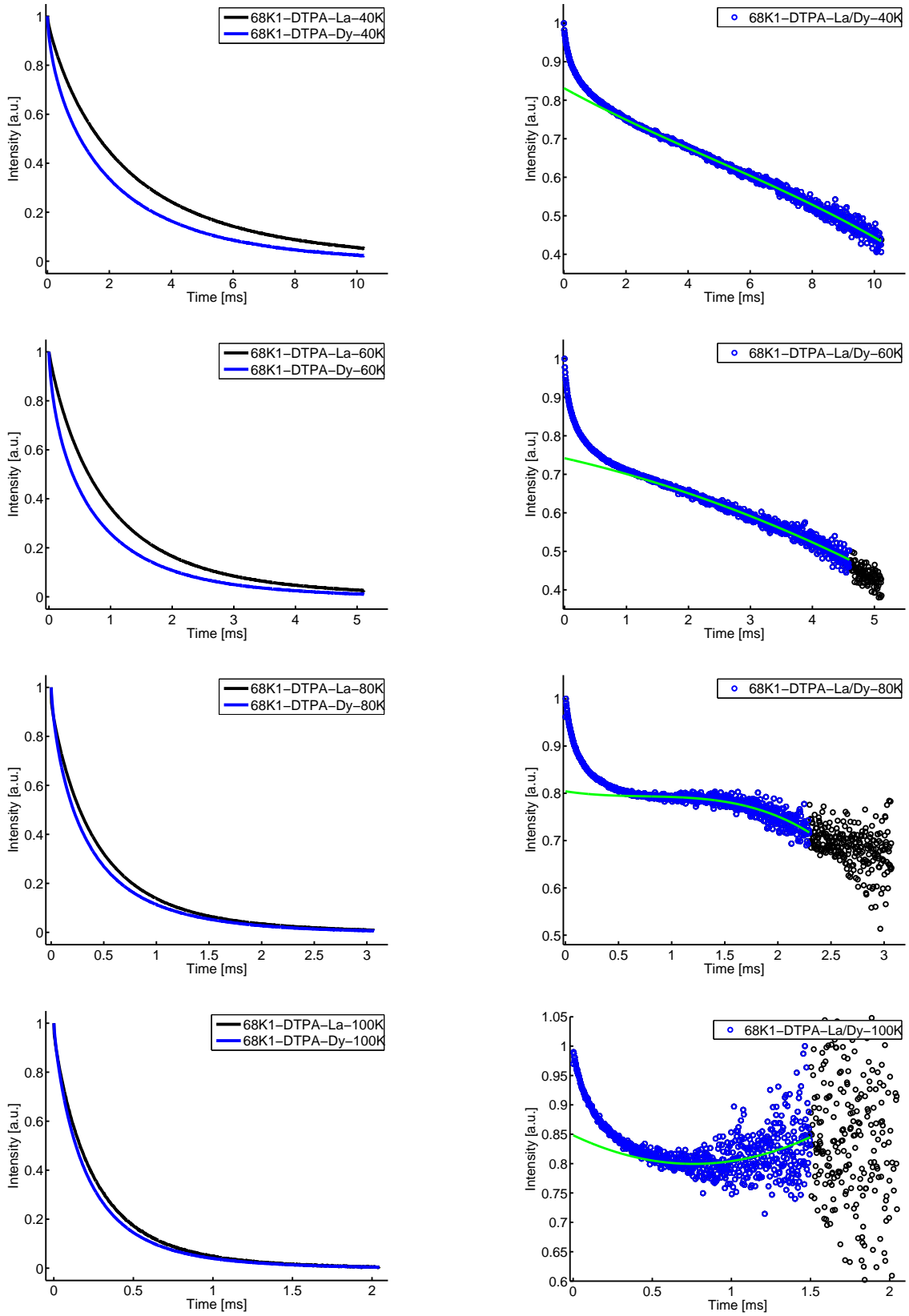


Figure S2: Relaxation data for sample 68K1-109[Dy(DTPA)] for temperatures $T=\{40\text{K}, 60\text{K}, 80\text{K}, 100\text{K}\}$. Left column of figures: Renormalized inversion recovery traces (Dy - blue, Lu - black). Right column of figures: Division of inversion recovery traces Dy/Lu and background fits (green). RE data points cutted out before background fitting are shown in black.

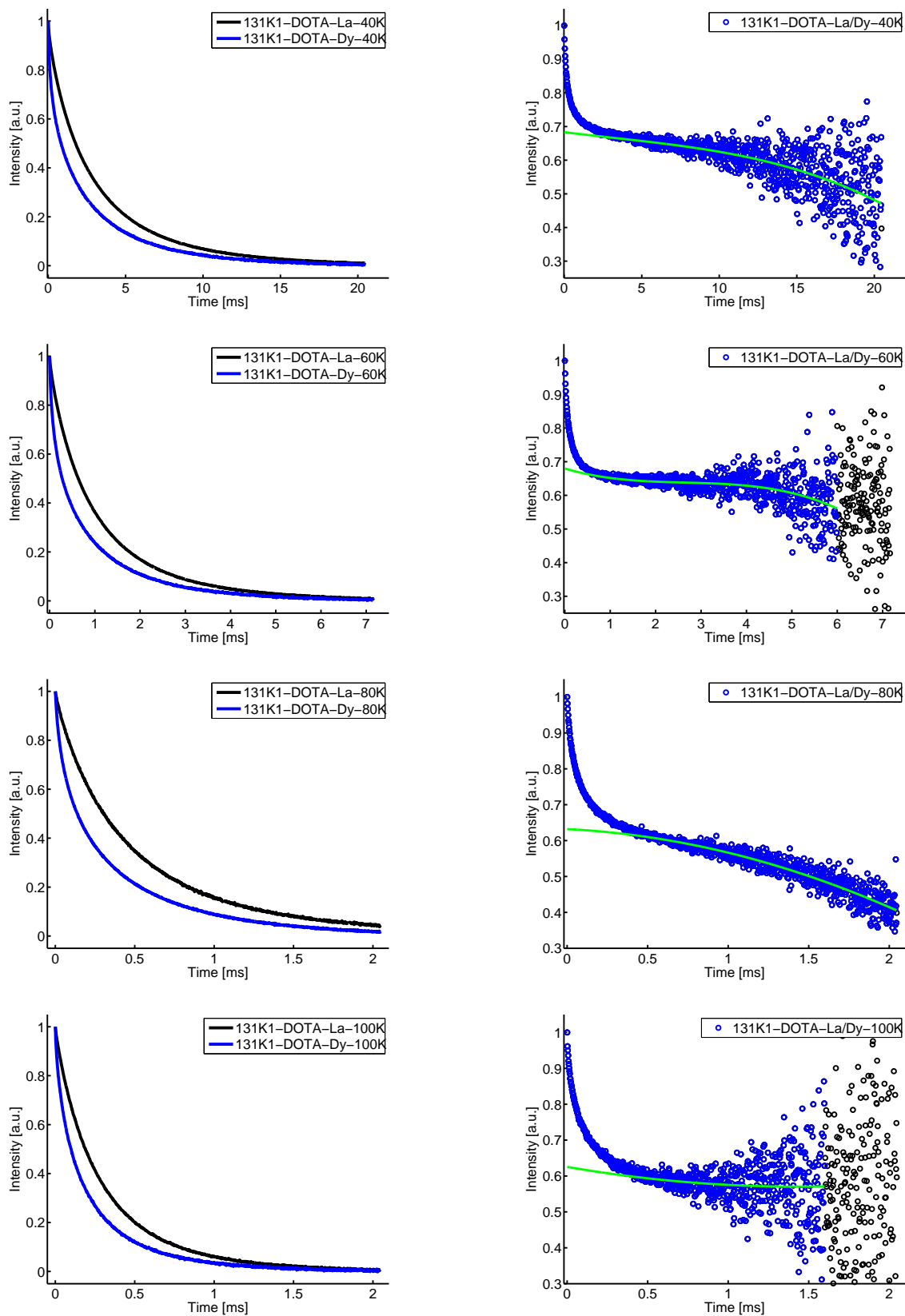


Figure S3: Relaxation data for sample 131K1-109[Dy(DOTA)] for temperatures $T=\{40\text{K}, 60\text{K}, 80\text{K}, 100\text{K}\}$. Left column of figures: Renormalized inversion recovery traces (Dy - blue, Lu - black). Right column of figures: Division of inversion recovery traces Dy/Lu and background fits (green). RE data points cutted out before background fitting are shown in black.

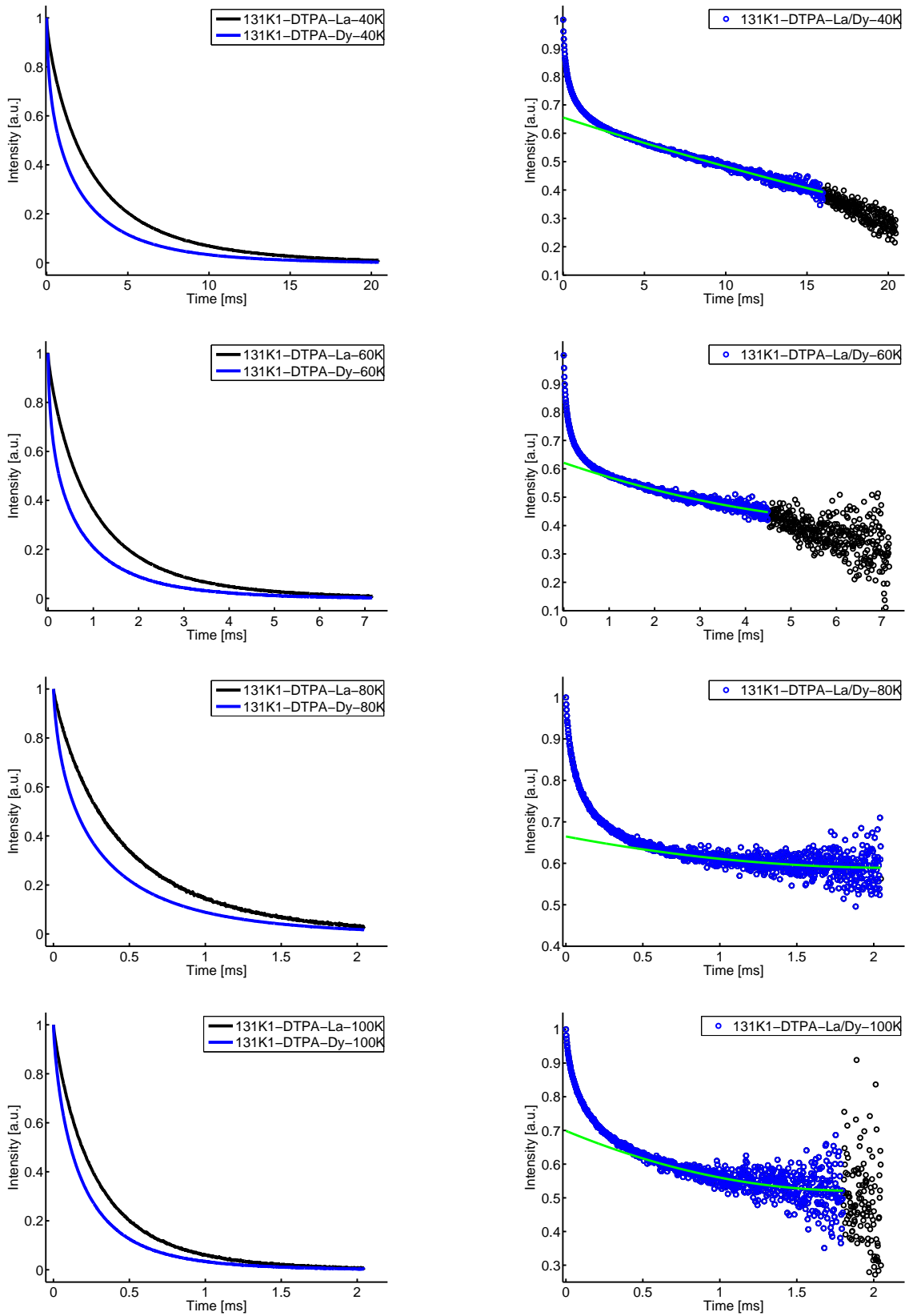


Figure S4: Relaxation data for sample 131K1-109[Dy(DTPA)] for temperatures $T=\{40\text{K}, 60\text{K}, 80\text{K}, 100\text{K}\}$. Left column of figures: Renormalized inversion recovery traces (Dy - blue, Lu - black). Right column of figures: Division of inversion recovery traces Dy/Lu and background fits (green). RE data points cutted out before background fitting are shown in black.