

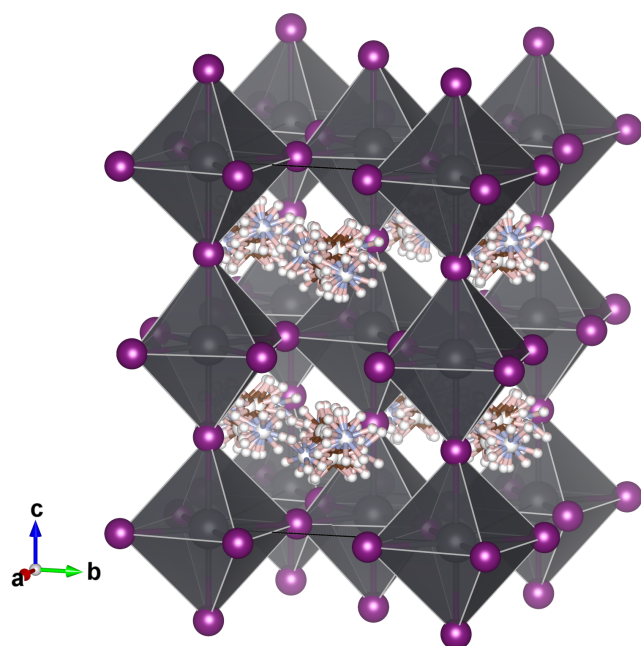
# Supporting Information to: Structural Investigations of $\text{MA}_{1-x}\text{DMA}_x\text{PbI}_3$ mixed-cation perovskites

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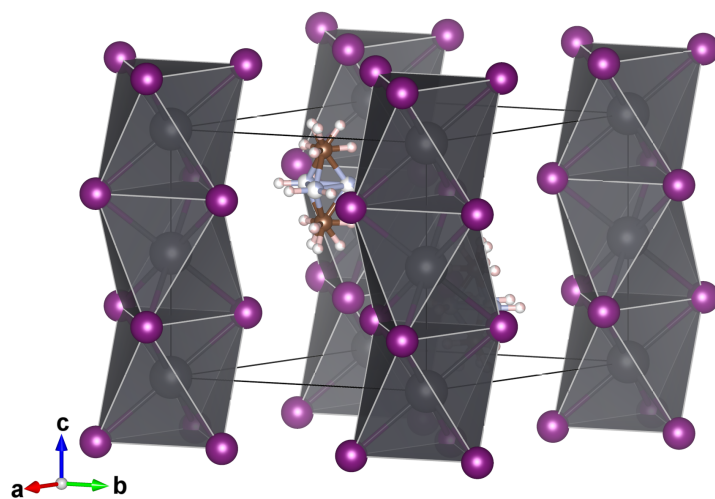
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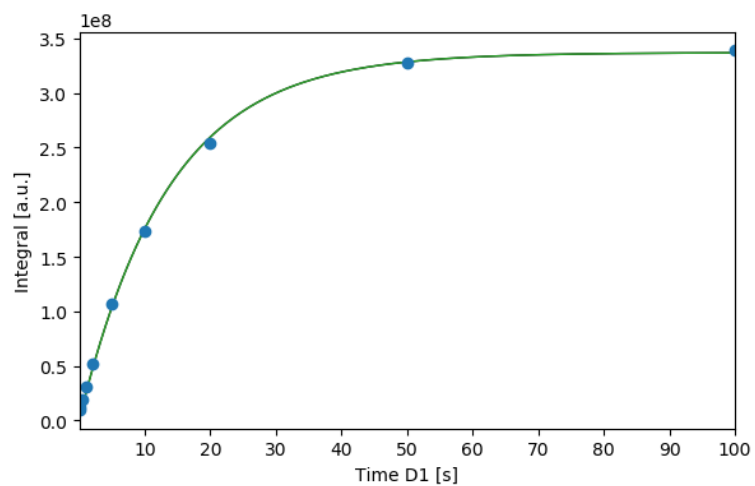
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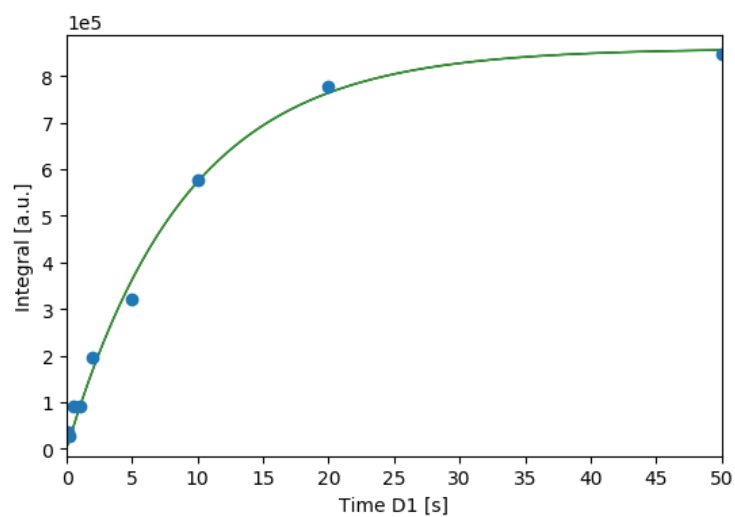
**Figure S1:** Room temperature crystal structure of  $\text{MAPbI}_3$ .



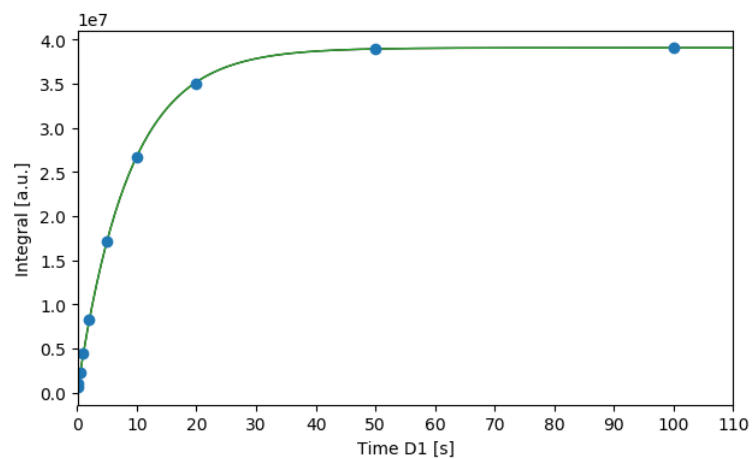
**Figure S2:** Room temperature crystal structure of  $\text{DMAPbI}_3$ .



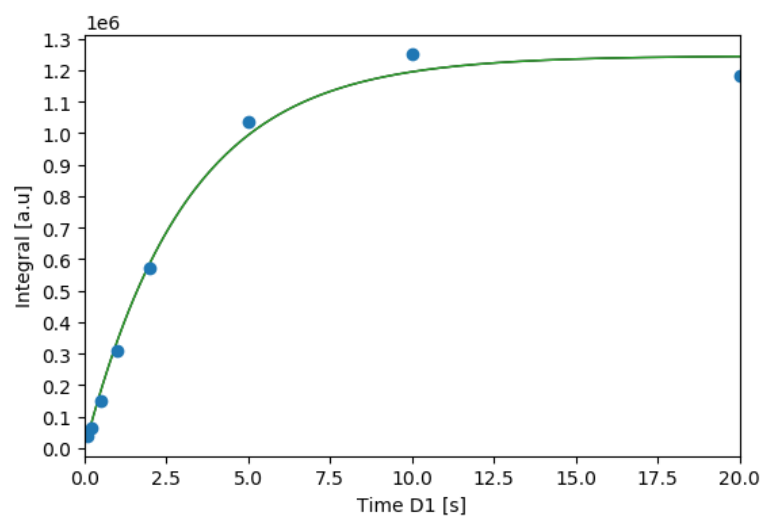
**Figure S3:**  $^1\text{H}$  saturation recovery  $T_1$  measurement of  $\text{MAPbI}_3$ . Measured at 20 T and 22 kHz MAS.



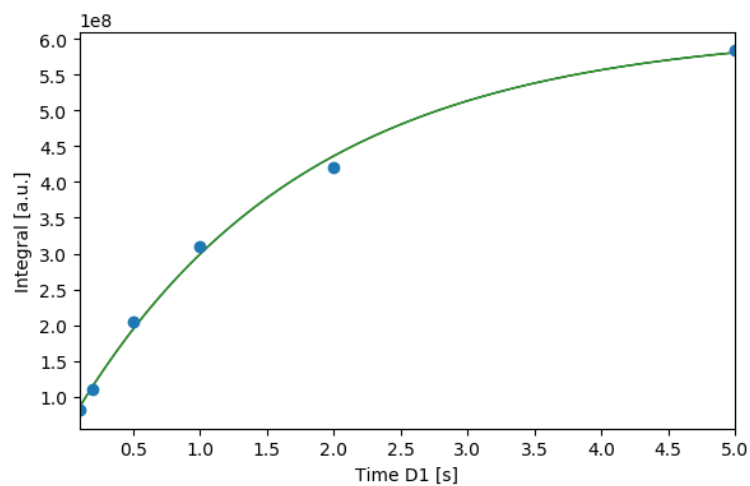
**Figure S4:**  $^{13}\text{C}$  saturation recovery  $T_1$  measurement of  $\text{MAPbI}_3$ . Measured at 20 T, 80 kHz  $^1\text{H}$  decoupling, and 22 kHz MAS.



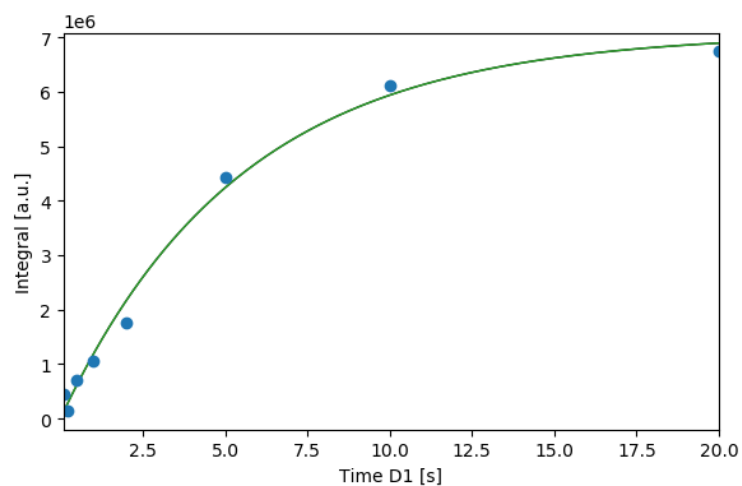
**Figure S5:**  $^1\text{H}$  saturation recovery  $T_1$  measurement of  $\text{DMAPbI}_3$ . Measured at 20 T and 20 kHz MAS.



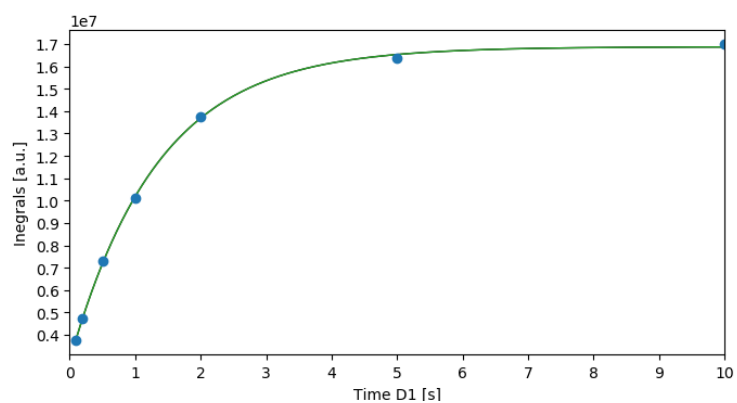
**Figure S6:**  $^{13}\text{C}$  saturation recovery  $T_1$  measurement of  $\text{DMAPbI}_3$ . Measured at 20 T, 80 kHz  $^1\text{H}$  decoupling, and 20 kHz MAS.



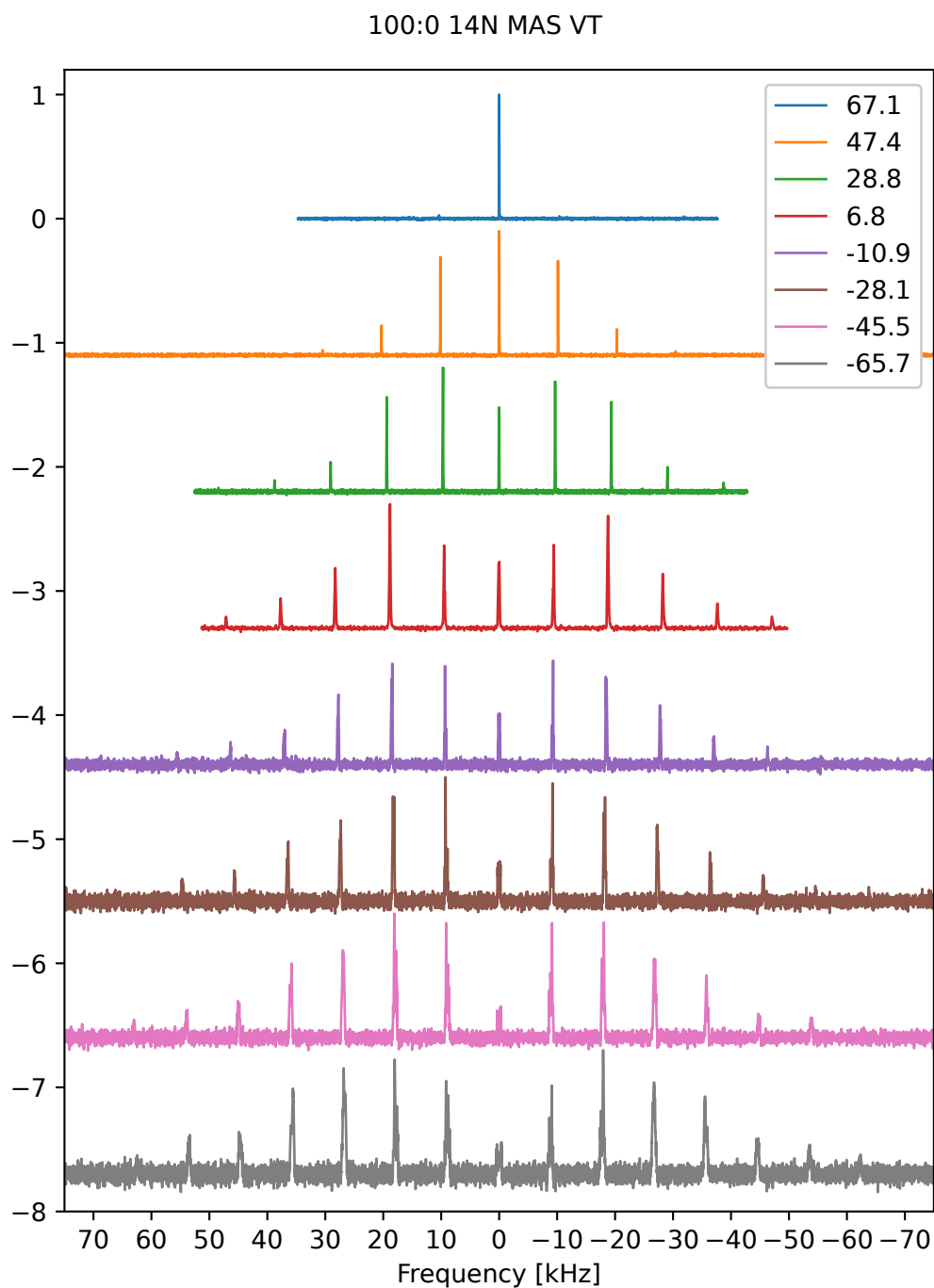
**Figure S7:**  $^{207}\text{Pb}$  saturation recovery  $T_1$  measurement of  $\text{DMApbi}_3$ . Measured at 20 T and using no MAS (static).



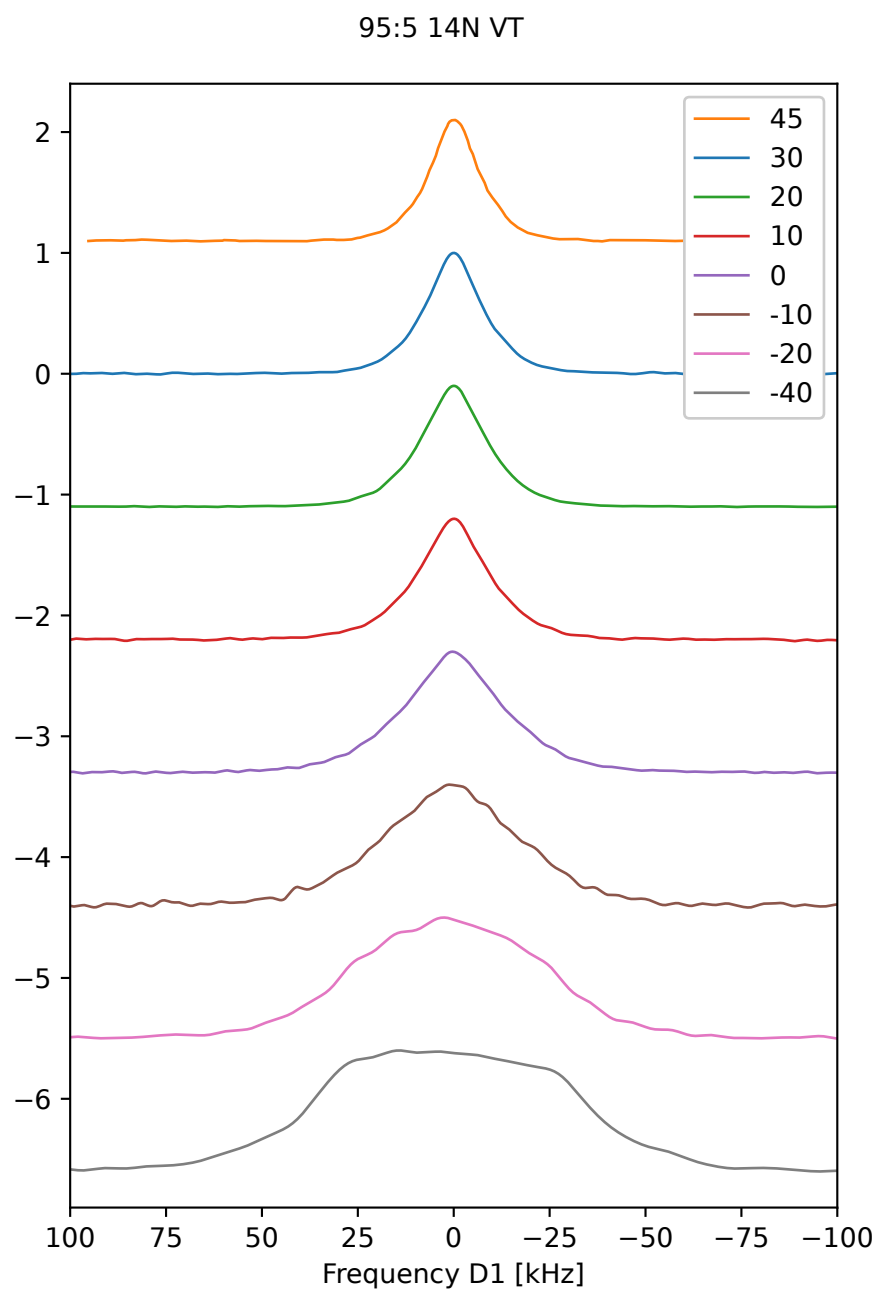
**Figure S8:**  $^{13}\text{C}$  saturation recovery  $T_1$  measurement of DMA in  $\text{MAPbI}_3$ . Measured at 20 T, 80 kHz  $^1\text{H}$  decoupling, and 17.5 kHz MAS.



**Figure S9:**  $^{207}\text{Pb}$  saturation recovery  $T_1$  measurement of the 95:5 sample. Measured at 20 T and using no MAS (static).

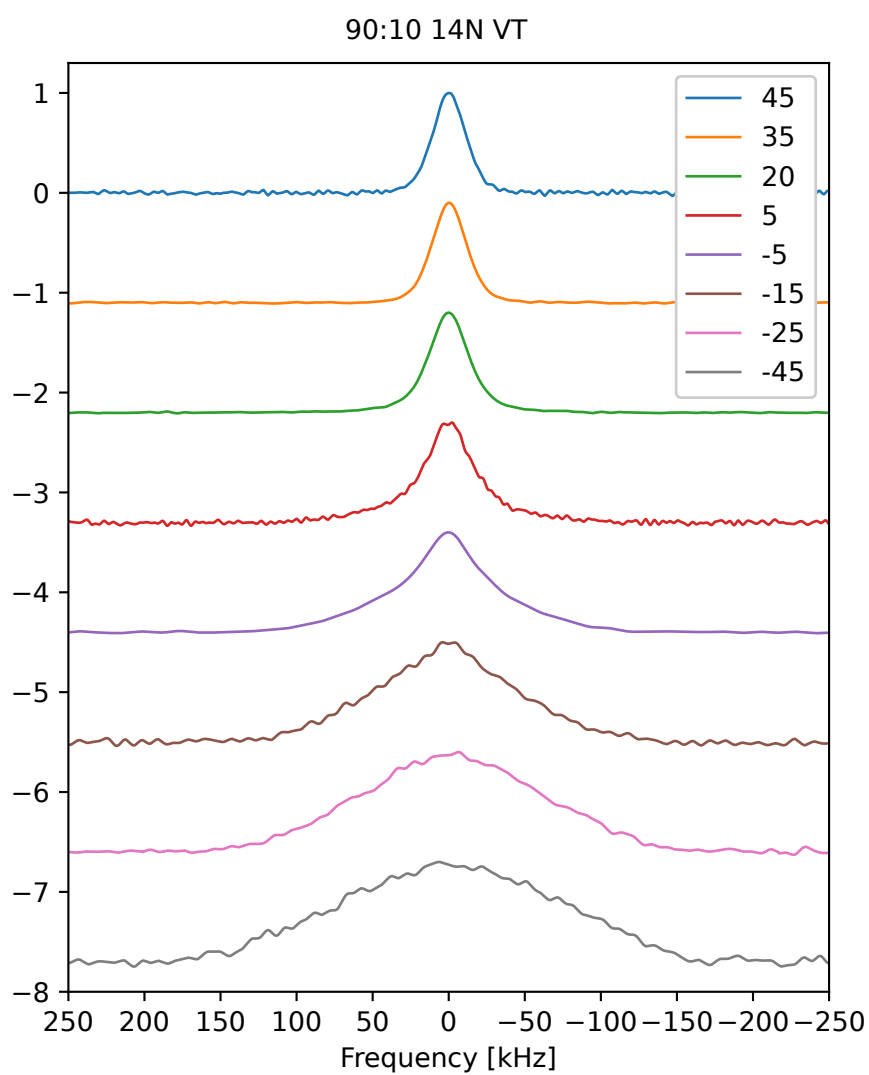


**Figure S10:**  $^{14}\text{N}$  MAS spectra of  $\text{MAPbI}_3$  (100:0 sample) at various temperatures ( $^{\circ}\text{C}$ ), measured at 9.4 T and using 9 kHz MAS. The integrals of the sidebands have been fit (assuming  $\eta = 0$ ) to obtain the  $C_Q$  as a function of temperature. Based on this value, the FWHM of the static line width has been estimated, using the distance between the maxima of the powder pattern.

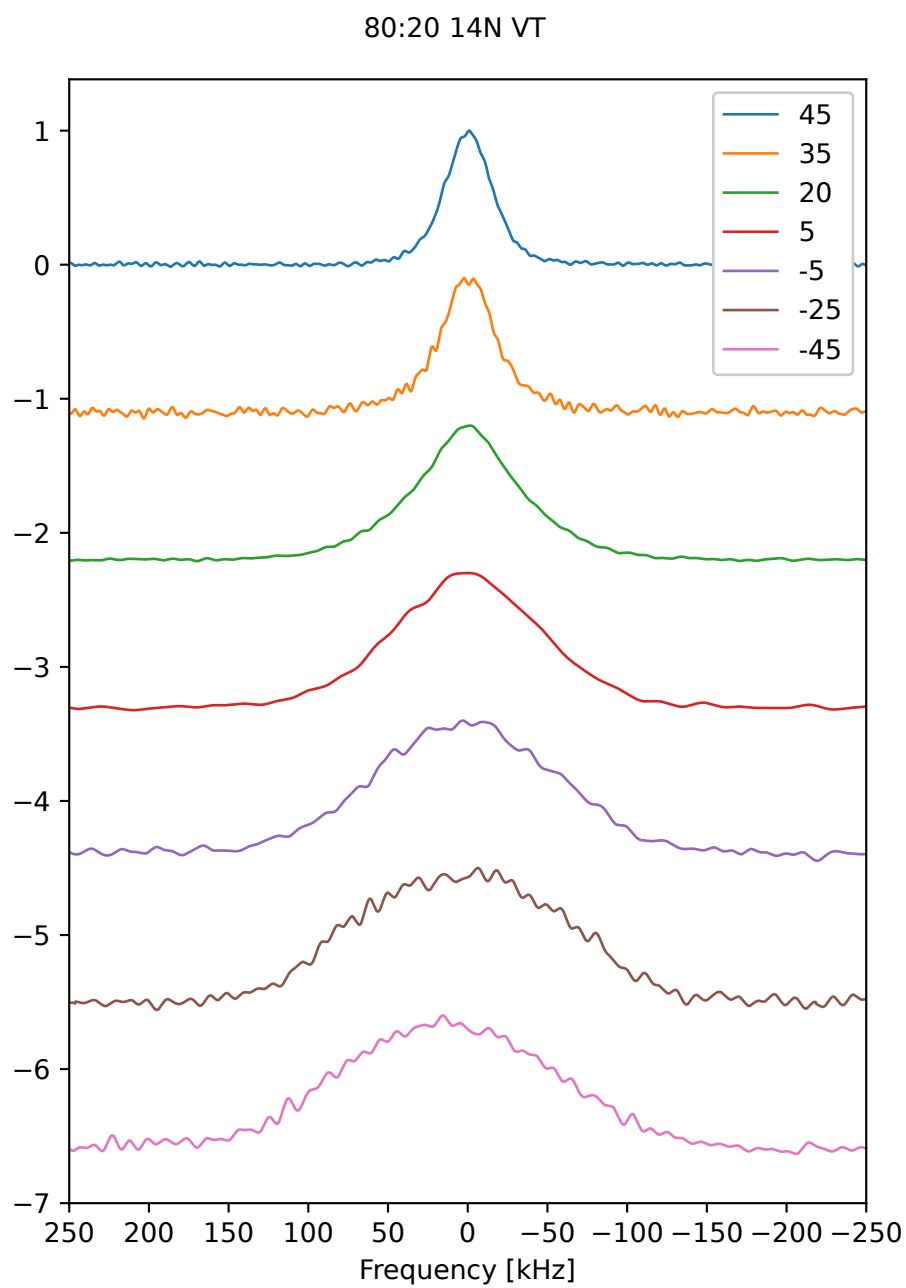


**Figure S11:** Static  $^{14}\text{N}$  spectra of (DMA:MA) $\text{PbI}_3$  (95:5 sample) at various temperatures ( $^{\circ}\text{C}$ ), measured at 20 T.

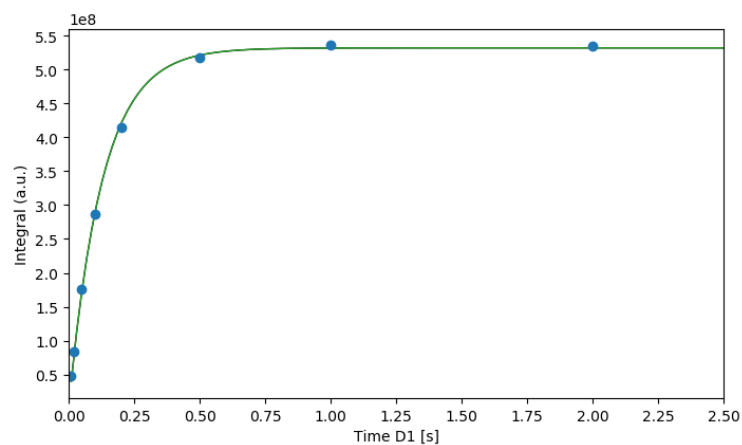




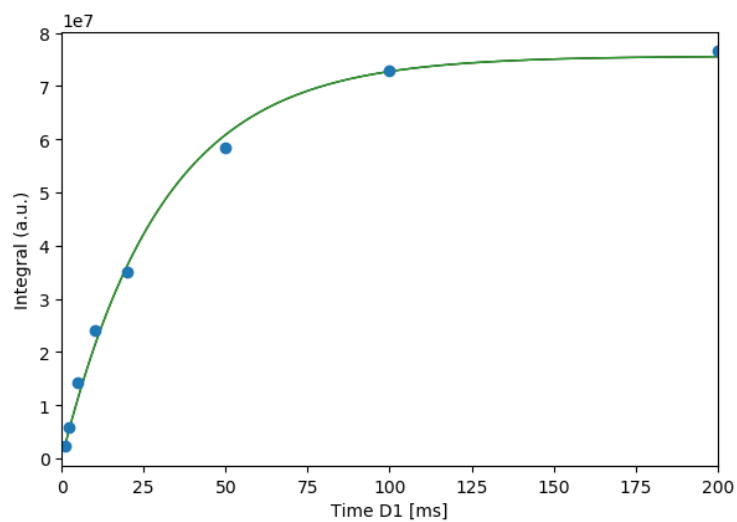
**Figure S12:** Static  $^{14}\text{N}$  spectra of (DMA:MA) $\text{PbI}_3$  (90:10 sample) at various temperatures ( $^{\circ}\text{C}$ ), measured at 20 T.



**Figure S13:** Static  $^{14}\text{N}$  spectra of (DMA:MA) $\text{PbI}_3$  (80:20 sample) at various temperatures ( $^{\circ}\text{C}$ ), measured at 20 T.



**Figure S14:**  $^{14}\text{N}$  saturation recovery  $T_1$  measurement of the MA cation in the 80:20 sample, used for establishing the reorientation time. The full curve shows the fit.



**Figure S15:**  $^{14}\text{N}$  saturation recovery  $T_1$  measurement of the DMA cation in the 80:20 sample, used for establishing the reorientation time. The full curve shows the fit.