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Supplementary Information to: Impact of magnetization and hyperfine field distribution on high magnetoelectric coupling strength in $BaTiO_3$ -BiFeO₃ multilayers

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Figure S1: a) - c) Ferroelectric P(V) hysteresis loops together with the I(V) characteristics of the three BaTiO₃-BiFeO₃ multilayer samples $15 \times BTO$ -BFO(50) (G5391b), $15 \times BTO$ -BFO(20) (G5392b), and $15 \times BTO$ -BFO(10) (G5393b). The loops were measured in a capacitor structure with the conducting SrTiO3:Nb substrate and 0.04 mm² Pt top contacts as electrodes. The maximum polarization increases with decreasing BFO thickness, except for the BTO-BFO(10) sample. Note that the contacts were selected randomly and are not identical with the contacts used for the magnetoelectric measurements. d) Photograph after voltage breakdown of the used contact The measurements were done in dynamic hysteresis mode with triangular excitation pulses at typically 1 kHz, except the thickest sample G5391 (2 kHz), and without leakage compensation.



Figure S2: For sample series $15 \times BTO$ -BFO(5) three measurements on the samples G5394b (@ 25 V voltage amplitude), and G5394d (@ 10 V and 19 V) are shown, with maximum polarization up to 48 C/cm². The measurements were done in dynamic hysteresis mode with triangular excitation pulses at typically 1 kHz, and without leakage compensation.



Figure S3: $2\theta - \omega$ scan of sample $15 \times BTO-BFO(50)$), with the SrTiO₃(001) substrate peak at 22.688°



Figure S4: 2 θ – ω scan of sample 15×BTO-BFO(20), with the SrTiO₃(001) substrate peak at 22.703^o



Figure S5: 2 θ - ω scan of sample 15×BTO-BFO(10), with the SrTiO₃(001) substrate peak at 22.688°



Figure S6: $2\theta-\omega$ scan of sample 15×BTO-BFO(5), with the SrTiO_3(001) substrate peak at 22.703^o



Figure S7: RSM of Sample $15 \times BTO-BFO(50)$ on the left the symmetric (001) direction and on the right the asymmetric ($\overline{1}03$) direction



Figure S8: RSM of Sample $15 \times BTO-BFO(20)$ on the left the symmetric (001) direction and on the right the asymmetric ($\overline{1}03$) direction



Figure S9: RSM of Sample $15 \times BTO-BFO(10)$ on the left the symmetric (001) direction and on the right the asymmetric ($\overline{1}03$) direction



Figure S10: RSM of Sample $15 \times BTO-BFO(5)$ on the left the symmetric (001) direction and on the right the asymmetric ($\overline{1}03$) direction