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Training the Polarization in Integrated La$_{0.15}$Bi$_{0.85}$FeO$_3$-Based Devices

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1 Derivation of $P_{\text{IP}}^\text{net}$

The polarization of BiFeO$_3$ projected along [001]$_{\text{p.c.}}$ is 60 $\mu$C/cm$^2$.[1, 2, 3, 4] As it is oriented along the pseudo-cubic ⟨111⟩ direction, the polarization magnitude can be calculated as

$$P_{[111]}(\text{BiFeO}_3) = \sqrt{3 \cdot P_{[001]}^2} = 104 \, \mu\text{C/cm}^2. \quad (1)$$

The single-domain in-plane polarization $P_{[110]}(\text{BiFeO}_3)$ is

$$P_{[110]}(\text{BiFeO}_3) = \sqrt{2 \cdot P_{[001]}^2} = 85 \, \mu\text{C/cm}^2. \quad (2)$$

$P_{\text{IP}}^\text{net}$ consists of a superposition of two domains with their in-plane polarizations rotated by 90°. We therefore derive it as

$$P_{\text{IP}}^\text{net} = P_{[100]}(\text{BiFeO}_3) = \frac{\sqrt{2 \cdot P_{[110]}^2}}{2} = 60 \, \mu\text{C/cm}^2. \quad (3)$$

2 SHG fit parameters

The non-zero parameters used for our SHG fits are displayed in Table 1. The obtained $\chi^{(2)}$-components are normalized to their maximum in order to compare their contribution to the tensor among the different samples and regions while omitting the need to account for differences in the total SHG yield. Optical SHG analysis on BiFeO$_3$ has been reported previously.[5] Note that possible strain-induced domain wall contributions were not identified.[6]
Table S1: Non-zero $\chi^{(2)}$-components obtained from our fits of the SHG anisotropy measurements in Figures 1, 2 and 3. The $\chi^{(2)}_{xxx}$-component (highlighted row) is supposed to be 0 for the point-group symmetry $m$.\textsuperscript{[7]}

| $\chi^{(2)}$-component | BiFeO$_3$ pristine | La$_{0.15}$Bi$_{0.85}$FeO$_3$ poled | La$_{0.15}$Bi$_{0.85}$FeO$_3$ poled capacitor |
|------------------------|------------------|-----------------------------|-----------------------------|
|                        | Magnitude | Phase | Magnitude | Phase | Magnitude | Phase |
| xxx                    | 0         | 0     | 0.20      | $\pi/2$ | 0         | 0     |
| xxy                    | 0.66      | 0     | 0.48      | $\pi/2$ | 0.48      | $\pi/4$ | 0.48 | 0     |
| yyy                    | 1         | $\pi$ | 1         | 3$\pi/4$ | 1         | 3$\pi/4$ | 1    | 3$\pi/4$ |
| yxx                    | 0.41      | 0     | 0.38      | 0      | 0.3       | 0     | 0.29 | $\pi/2$ |

3 Scanning-probe microscopy of BiFeO$_3$ and La$_{0.15}$Bi$_{0.85}$FeO$_3$

Figure S1: Same-scale AFM (a, b) and lateral-PFM (c, d) images of BiFeO$_3$ (a, c) and La$_{0.15}$Bi$_{0.85}$FeO$_3$ (b, d).
4 Negatively poled state

Figure S2: SHG images of the same capacitor in positively (left) and negatively (right) poled state.
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