Supplementary Information (SI) for Nanoscale Horizons.

Supporting Information Appendix

Advancing Green Electronics: Tunable Piezoelectric Enhancement in Biodegradable Poly (L-Lactide Acid) PLLA Films through Thermal-Strain Engineering

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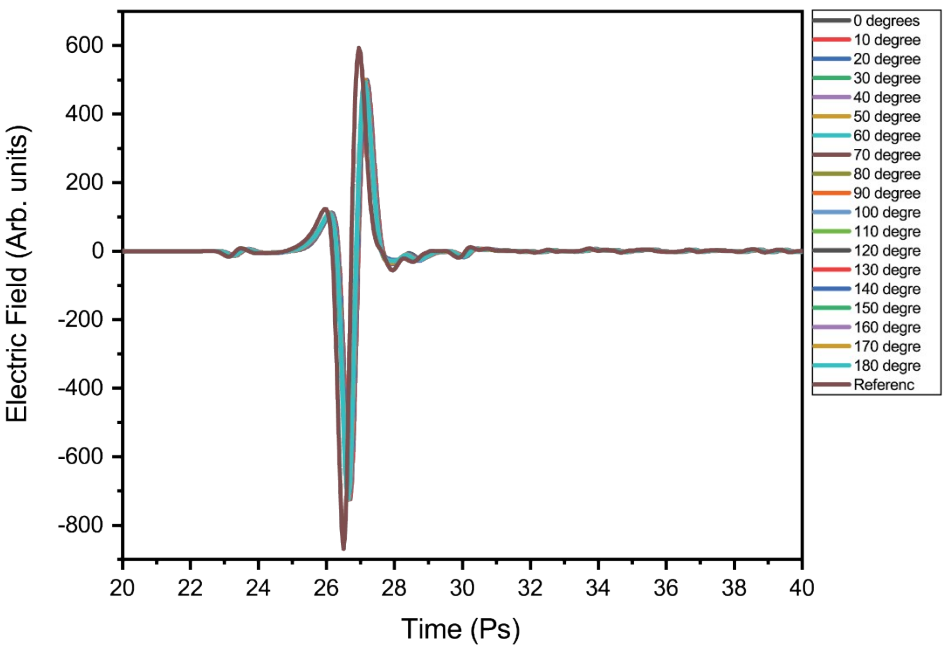
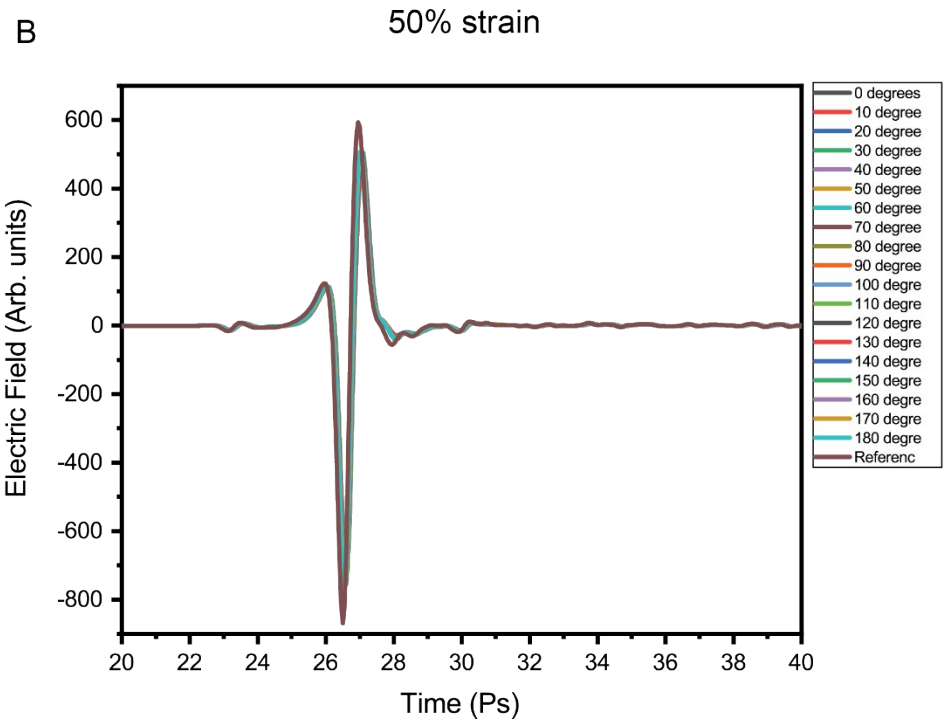
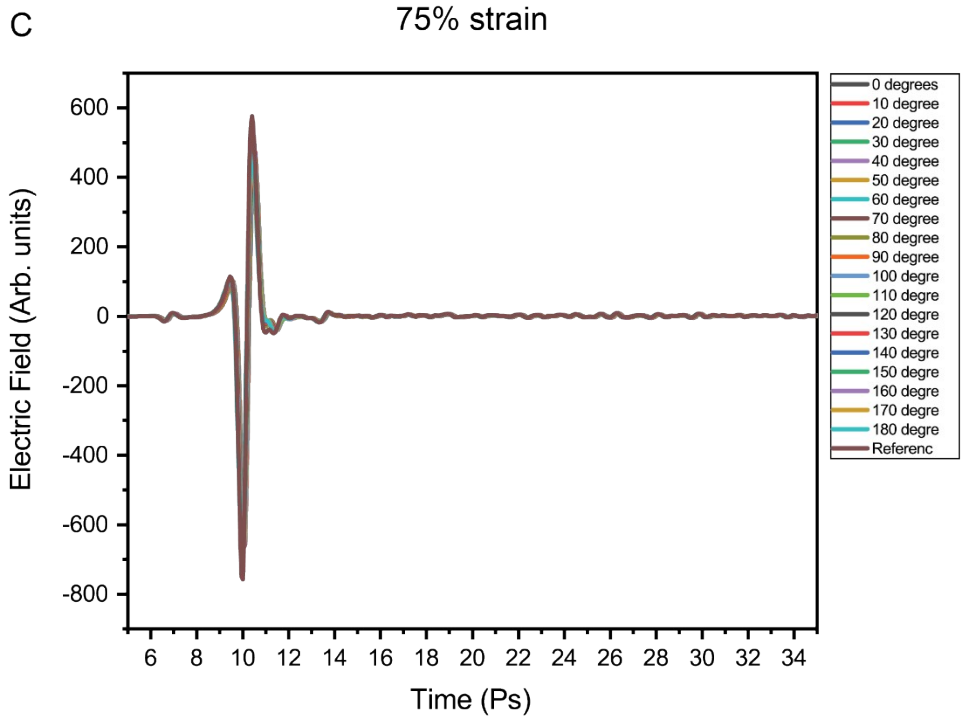
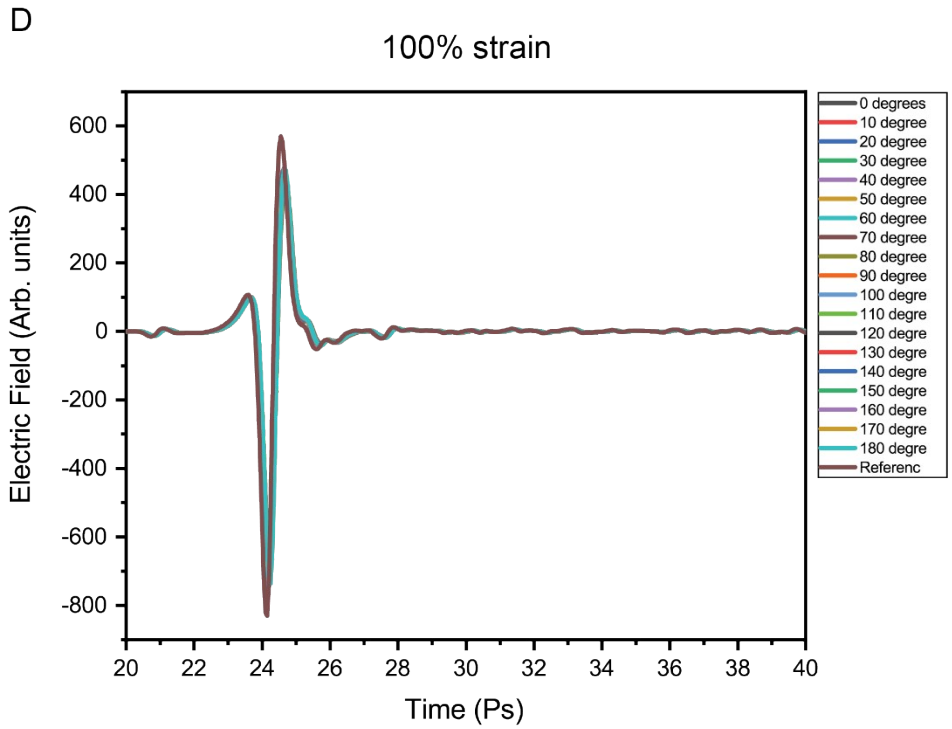
A


Unprocessed
Electric Field (Arb. units)
Time (Ps)
0 degrees
10 degree
20 degree
30 degree
40 degree
50 degree
60 degree
70 degree
80 degree
90 degree
100 degre
110 degre
120 degre
130 degre
140 degre
150 degre
160 degre
170 degre
180 degre
Referenc

B

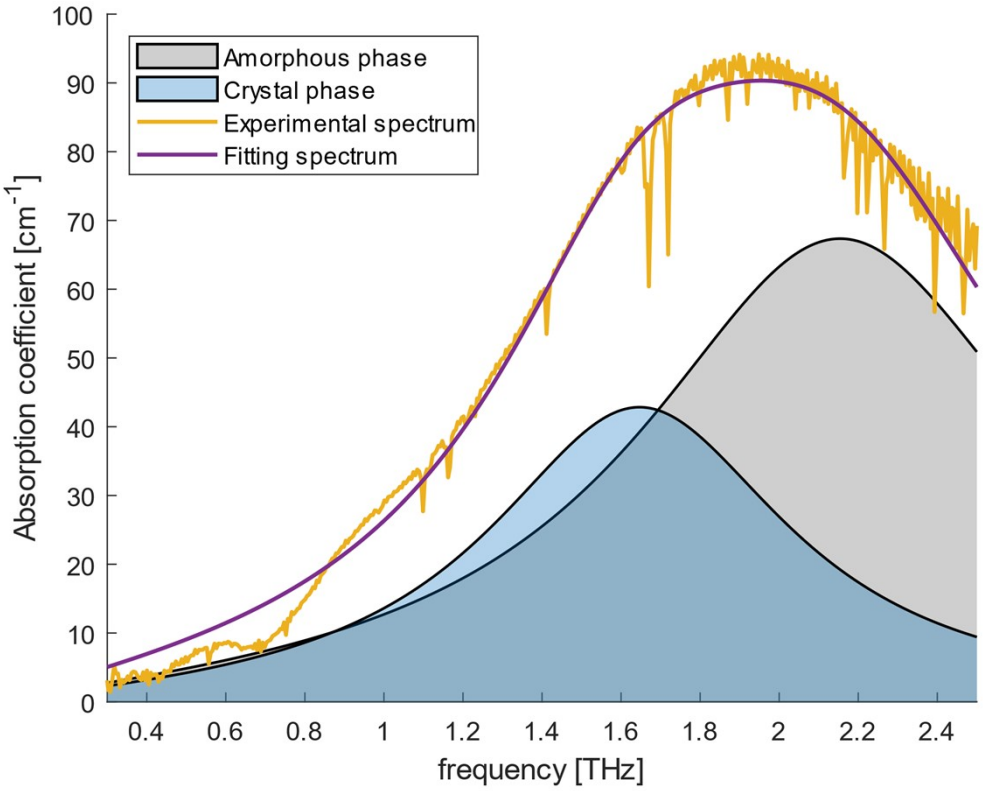

50% strain
Electric Field (Arb. units)
Time (Ps)
0 degrees
10 degree
20 degree
30 degree
40 degree
50 degree
60 degree
70 degree
80 degree
90 degree
100 degre
110 degre
120 degre
130 degre
140 degre
150 degre
160 degre
170 degre
180 degre
Referenc



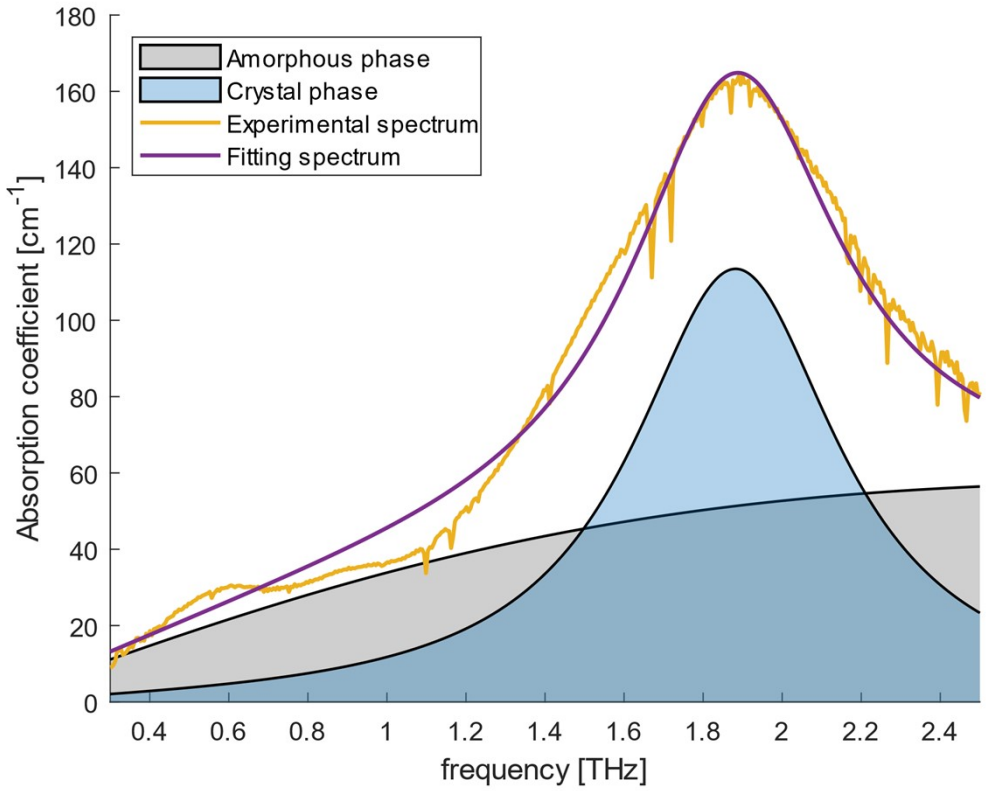


S 1 Terahertz wave transmission as a function of rotational angle (0-180°) for unprocessed, 50% strained, 75% strained, and 100% strained films. Each plot illustrates the effect of mechanical strain on the terahertz wave response as the film is rotated. The unprocessed film serves as the control, showing the baseline terahertz response, while increasing strain percentages (50%, 75%, 100%) demonstrate corresponding shifts in terahertz transmission, reflecting changes in molecular orientation and crystallinity.

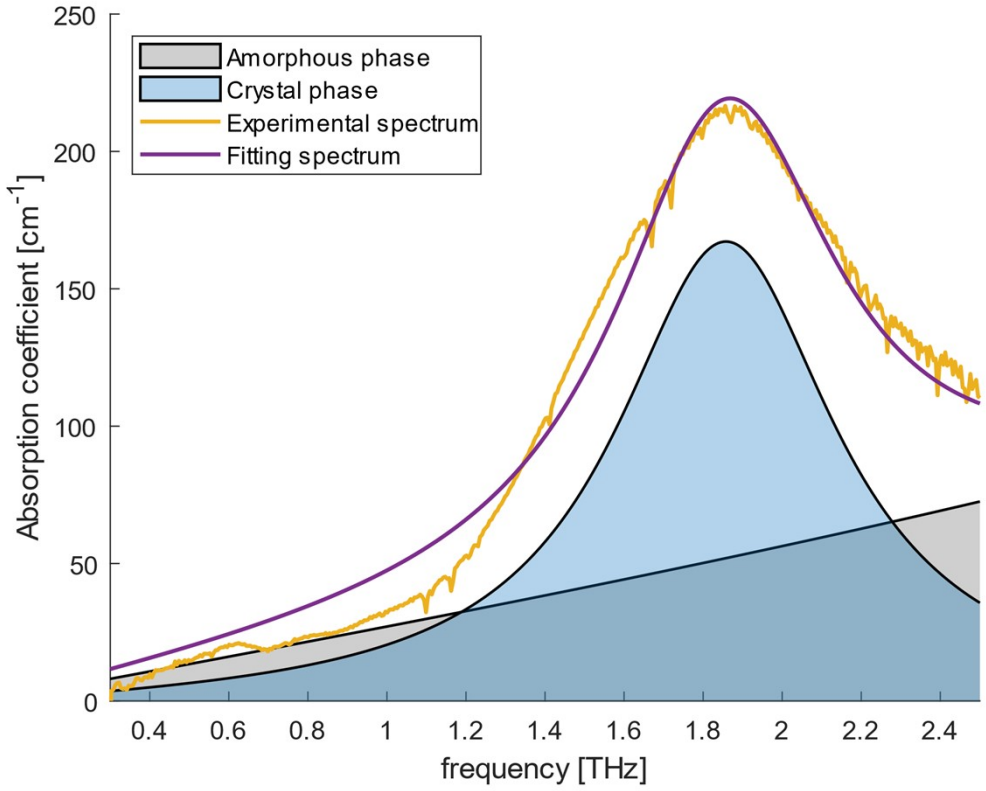
Unprocessed film

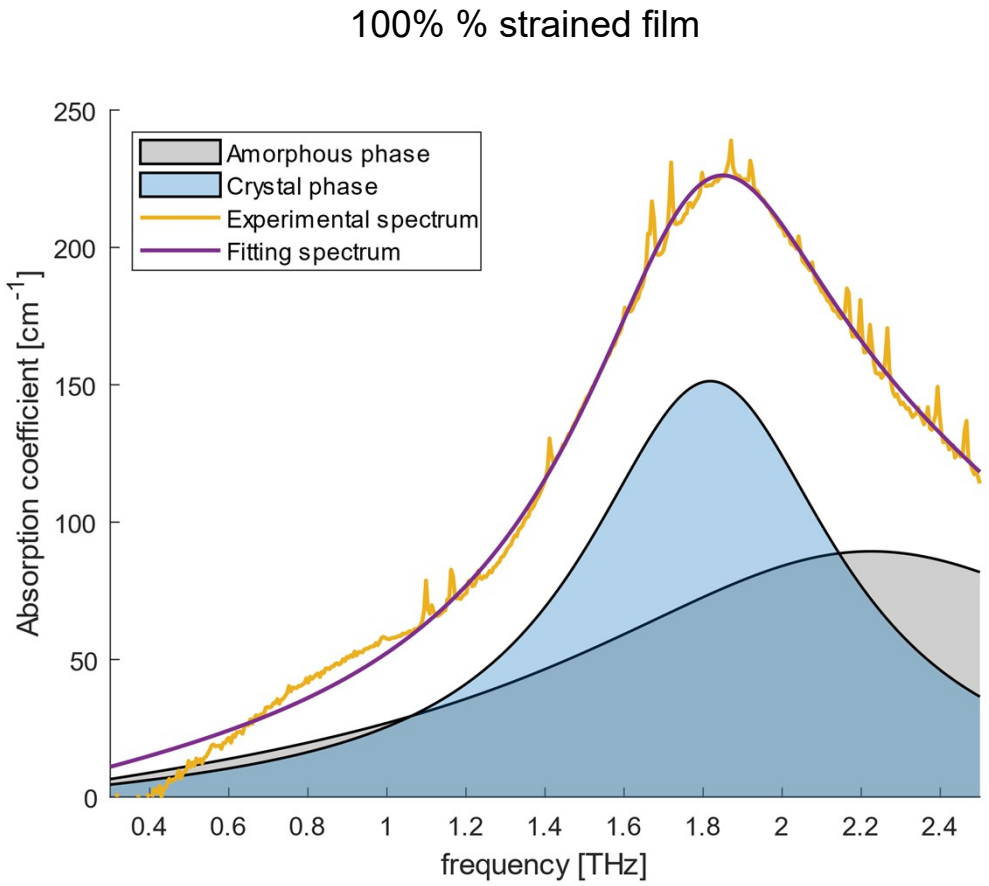


50% strained film



75% strained film


Amorphous phase
Crystal phase
Experimental spectrum
Fitting spectrum
Absorption coefficient [cm-1]
frequency [THz]
250
200
150
100
50
0
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
2
2.2
2.4



S 2 Analysis of the crystallinity quantification of PLLA films under different strain conditions, Unprocessed (A) 50% (B), 75% (C), and 100%(D), using THz-TDS. The yellow lines represent the experimental terahertz data, while the purple lines denote the fitted results. As the strain increases, changes in the terahertz response are observed, indicating variations in molecular orientation and crystallinity. The blue-shaded areas correspond to the crystalline contributions, while the grey areas represent the amorphous contributions. These responses result from the interplay between Lorentz oscillators, which describe the vibrational modes interacting with terahertz electromagnetic waves. The dissipation of the terahertz waves is proportional to the number of oscillators in each mode, represented by α_j*. As the area of* $\varepsilon''(\omega)$ *is proportional to* α_j *we use the area of* $\varepsilon''(\omega)$ *as an indicator of these contributions. The sum of the areas of* $\varepsilon''(\omega)$ *contributions from crystalline regions are defined as* I_{XC}*, and the area belonging to the amorphous phase is defined as* I_{XA} *allowing for a calculation of crystallinity* X_{CTHz} *based on the following equation:*

$$X_{CTHz} = \frac{I_{XC}}{I_{XC} + I_{XA}} \cdot 100$$

Piezoresponse force microscopy (PFM) of strained PLLA films.

1. PLLA 0%.

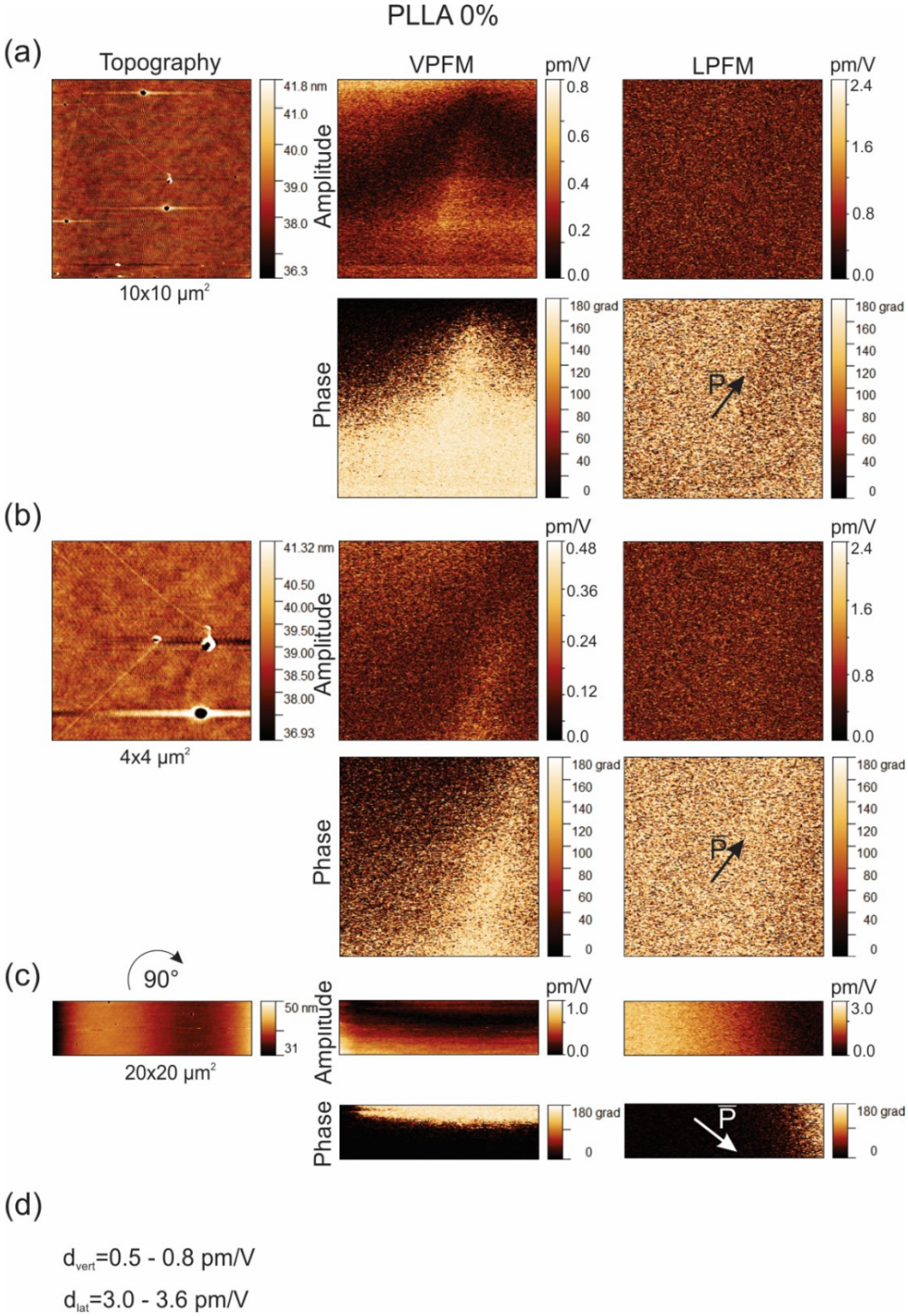


Figure. 1. Scanning probe microscopy of PLLA 0% film. (a), (b) Surface topography and the PFM (right) at different scales, 10x10 μm² and 4x4 μm². (c) Surface topography and the PFM (right) after azimuth angle rotation on 90°. (d) vertical VPFM and lateral LPFM effective riezocoefficient's values. PFM images were

obtained at a probe voltage of 4, 4 and 10 V for (a), (b) and (c) respectively. Lateral polarization is schematically marked by arrows in phase images.

2. PLLA 50%

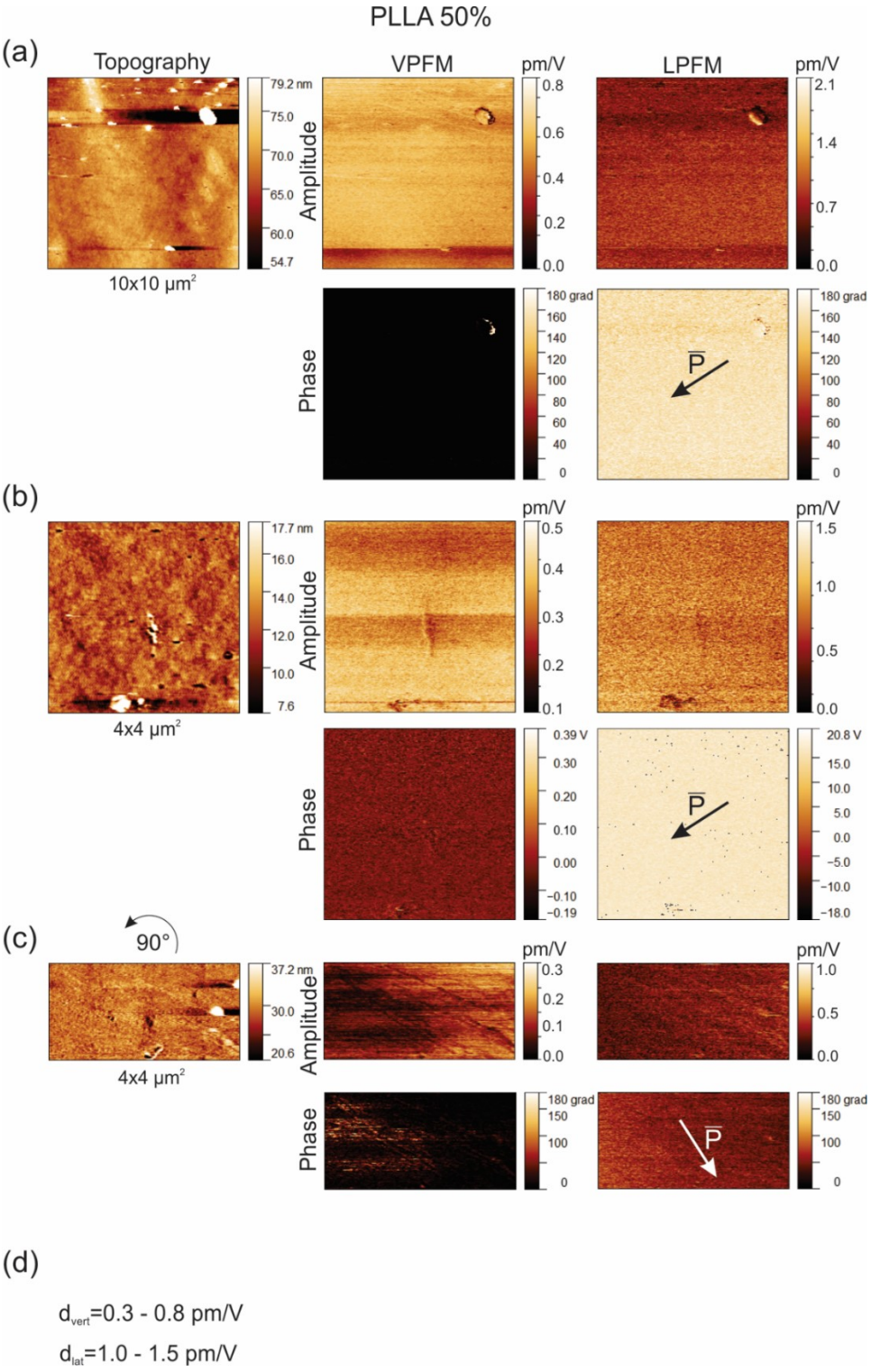


Figure. 2. Scanning probe microscopy of PLLA 50% film. (a), (b) Surface topography and the PFM (right) at different scales, 10x10 μm² and 4x4 μm². (c) Surface topography and the PFM (right) after azimuth angle rotation on 90°. (d) vertical VPFM and lateral LPFM effective riezocoefficient's values. PFM images were

obtained at a probe voltage of 20, 20 and 40 V for (a), (b) and (c) respectively. Lateral polarization is schematically marked by arrows in phase images.

3. PLLA 75%.

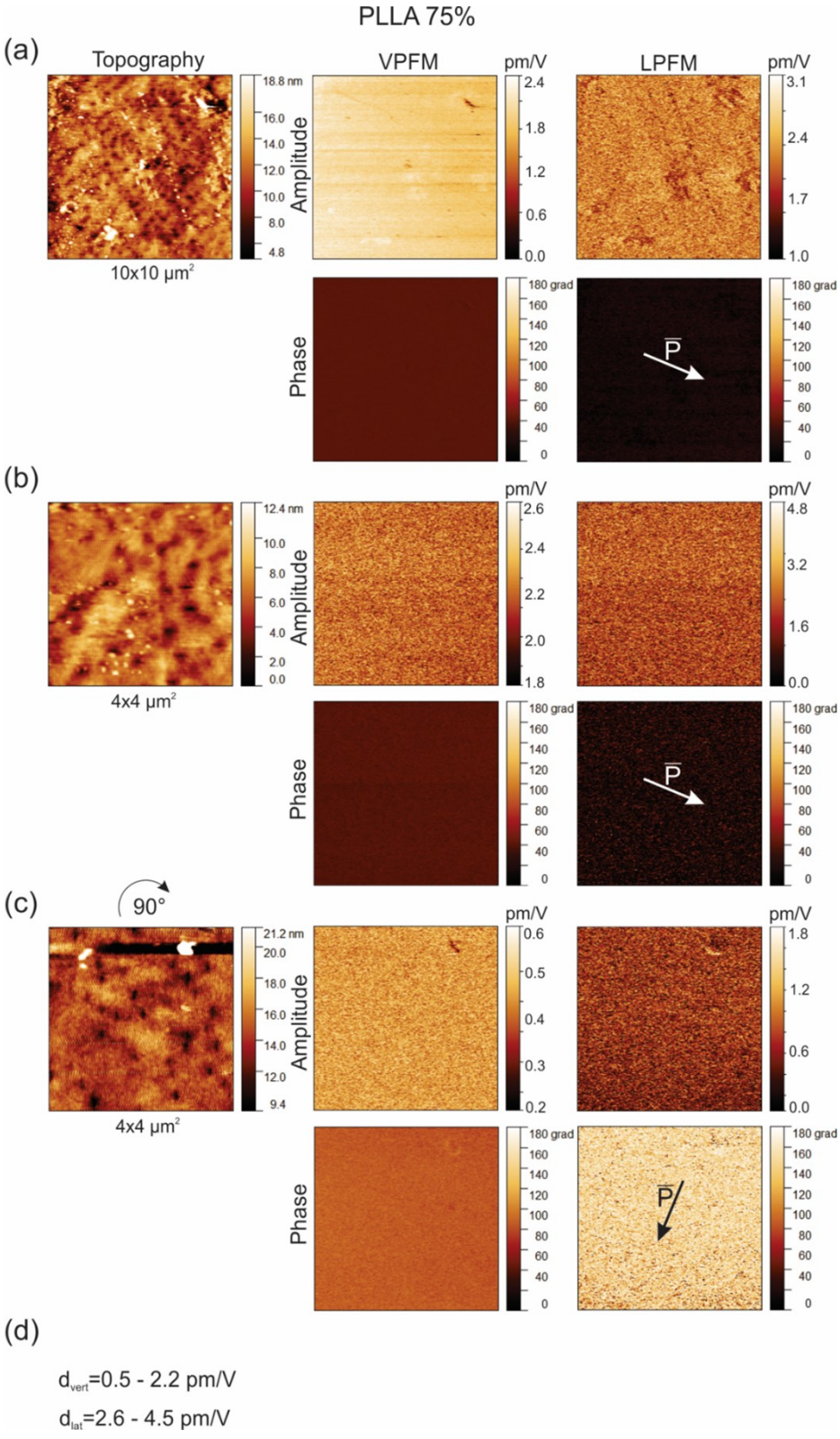


Figure. 3. Scanning probe microscopy of PLLA 75% film. (a), (b) Surface topography and the PFM (right) at different scales, 10x10 μm² and 4x4 μm². (c) Surface topography and the PFM (right) after azimuth angle rotation on 90°. (d) vertical VPFM and lateral LPFM effective riezocoefficient's values. PFM images were

obtained at a probe voltage of 16, 4 and 10 V for (a), (b) and (c) respectively. Lateral polarization is schematically marked by arrows in phase images.

4. PLLA 100%.

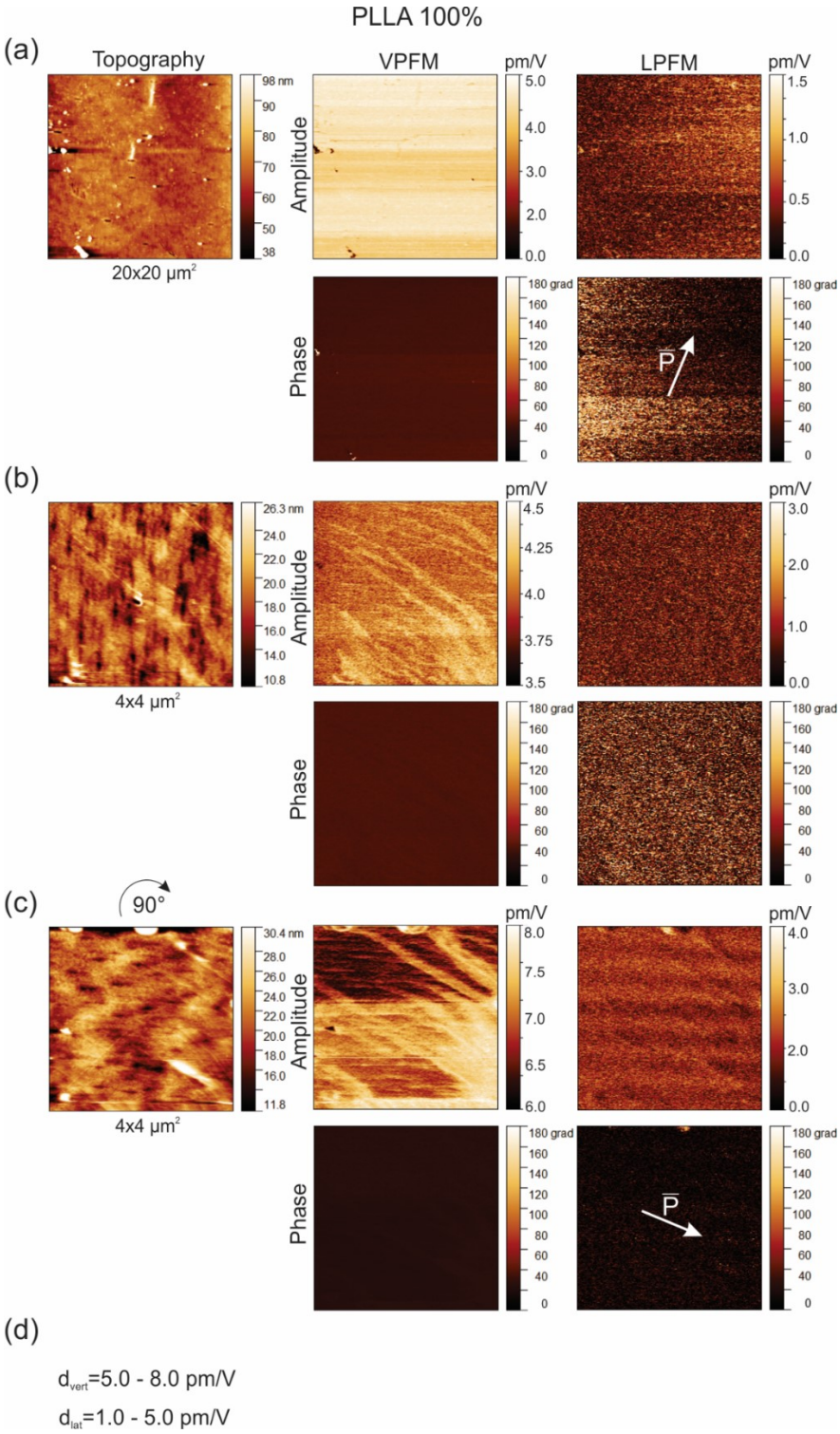


Figure. 4. Scanning probe microscopy of PLLA 100% film. (a), (b) Surface topography and the PFM (right) at different scales, 20x20 μm² and 4x4 μm². (c) Surface topography and the PFM (right) after azimuth angle rotation on 90°. (d) vertical VPFM and lateral LPFM effective riezocoefficient's values. PFM images were obtained at a probe voltage of 6, 4 and 6 V for (a), (b) and (c) respectively. Lateral polarization is schematically marked by arrows in phase images.