Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information

Decomposing and Analyzing Contact Resonance Frequency in Contact Mode Voltage Modulated Scanning Probe Microscopies

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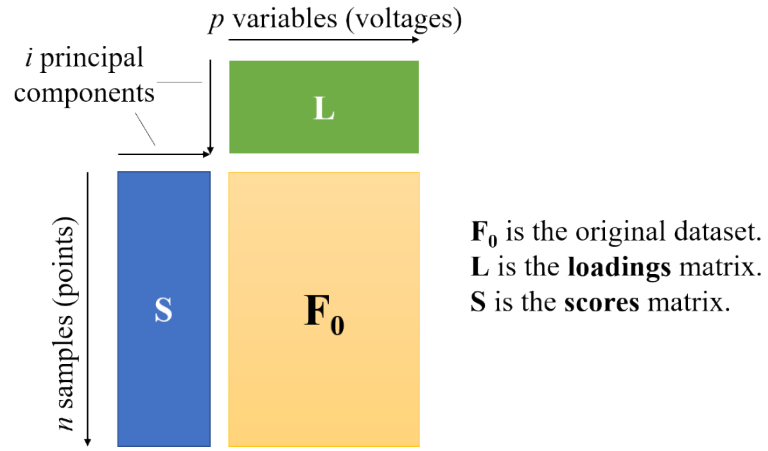

Fig. S1 The schematic diagram of principal component analysis (PCA).

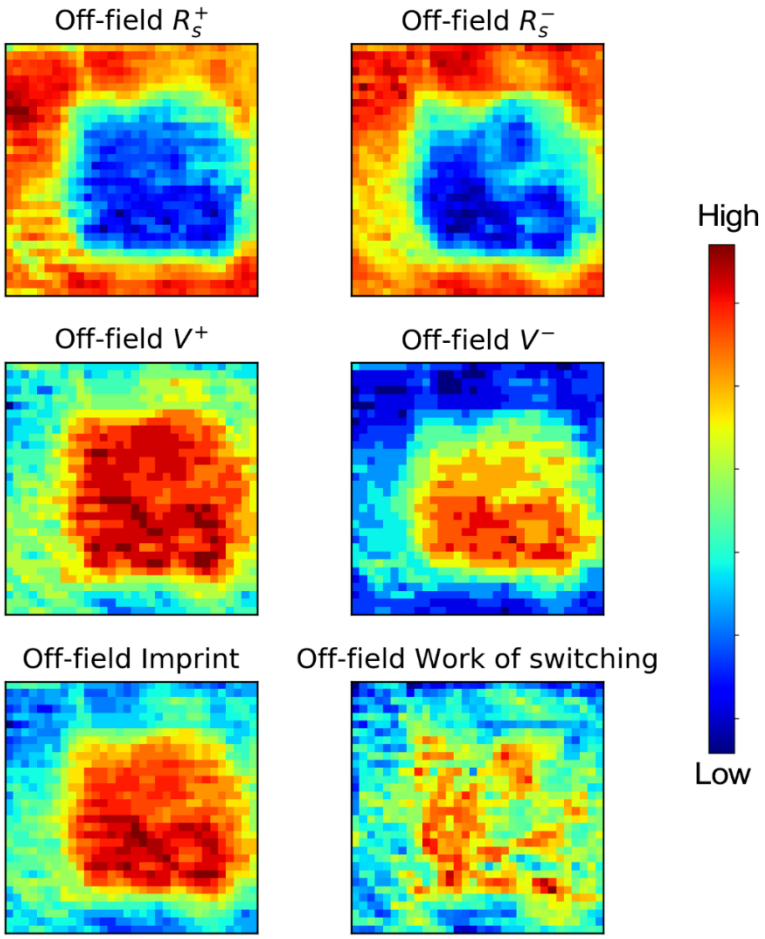

Fig. S2 The maps of switching parameters calculated from the SS-PFM measurements that were conducted on PZN-4.5%PT at on- and off-field. First row images: forward and reverse saturation responses, R_s^+, R_s^-; second row images: forward and reverse coercive voltages, V^+ and V^-; and third row images: the work of switching is defined as the area within the loop. The imprint bias is defined as $Im = (V^+ + V^-)/2$. Image area: 4 × 4 μm^2 with a resolution of 32 × 32 points in the same area as showed in Figure 2 of the main text. The off-field and on-field hysteresis loops are shown in the Fig. 1(c, f) of the main text of this paper, respectively.

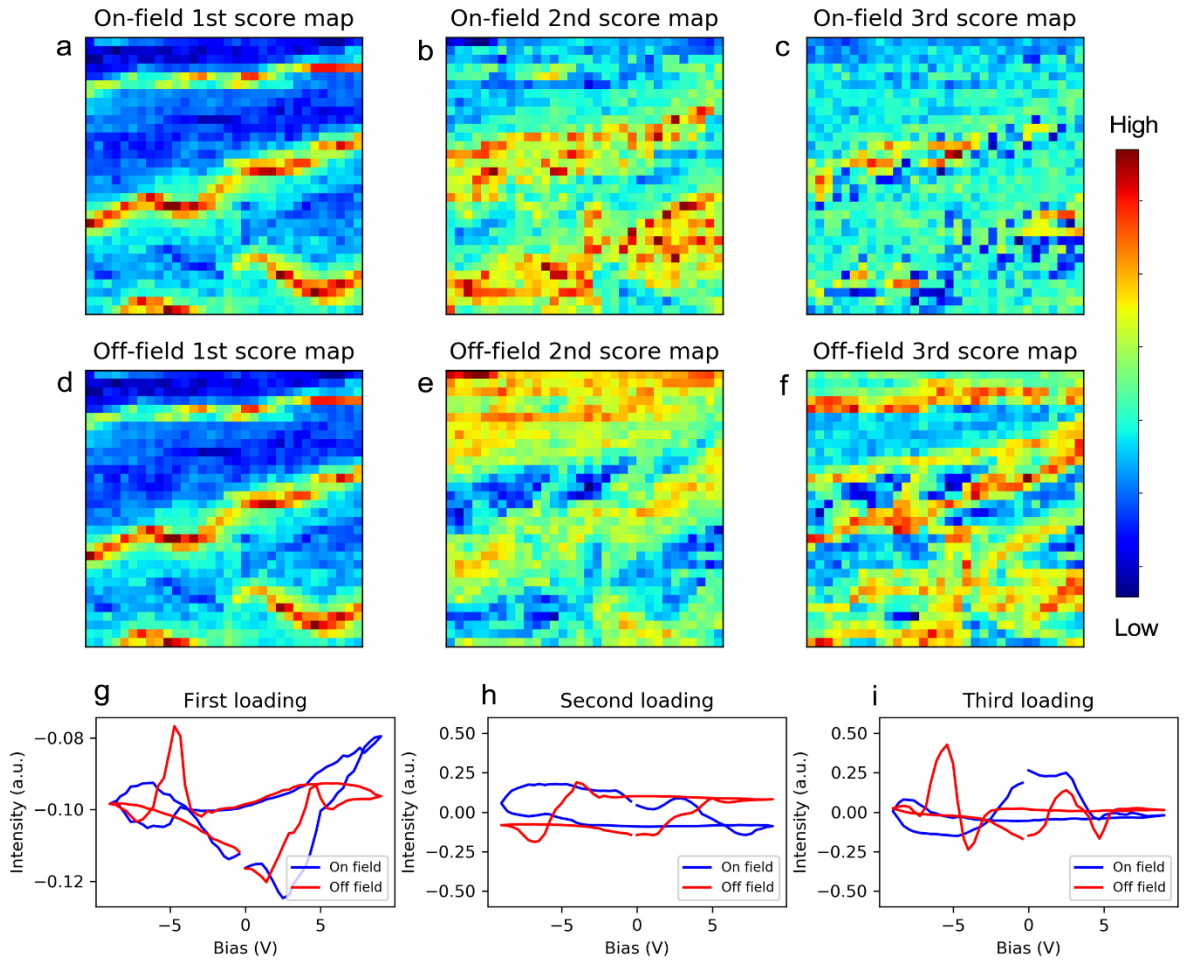

Fig. S3 Principal component analysis (PCA) on the contact resonance frequency (f_0) of PZN-4.5%PT in the SS-PFM measurements: (a-c) The score maps for the first three principal components at the on-field; (d-f) The score maps for the first three principal components at the off-field; and (g-i) The loadings for the first three principal components. The percentages of variance explained by the first, the second, and the third on-field principal components are 87.95%, 3.74%, and 1.20%, respectively; at the off field, they are 91.35%, 2.87%, and 0.85%, respectively. The SS-PFM mapping is scanned in a 4 × 4 μm^2 area with a resolution of 32 × 32 points. The area is with spontaneous polarization.

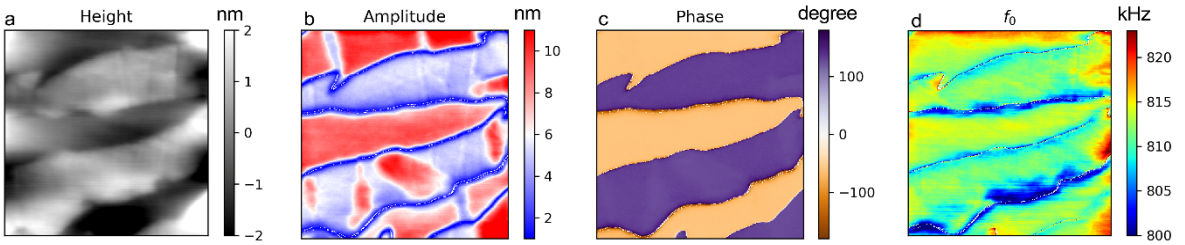

Fig. S4 The PFM images of PZN-4.5%PT: (a) Height; (b) Amplitude; (c) Phase; and (d) Contact resonance frequency (f_0) in an area of 4 × 4 μm^2. The resolution is 256 × 256 pixels. The area is with spontaneous polarization.

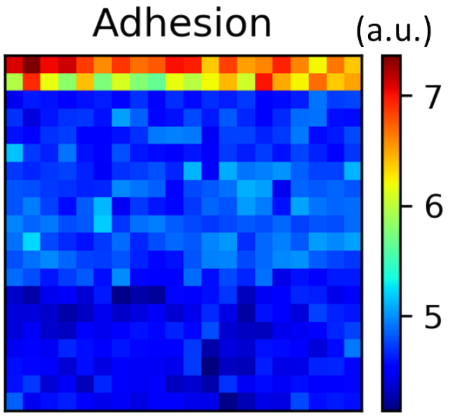

Fig. S5 The adhesion force map in the SM-ESM measurement conducted on PMMA. The maps are scanned in a 10 × 10 μm^2 area with a resolution of 20 × 20 points.

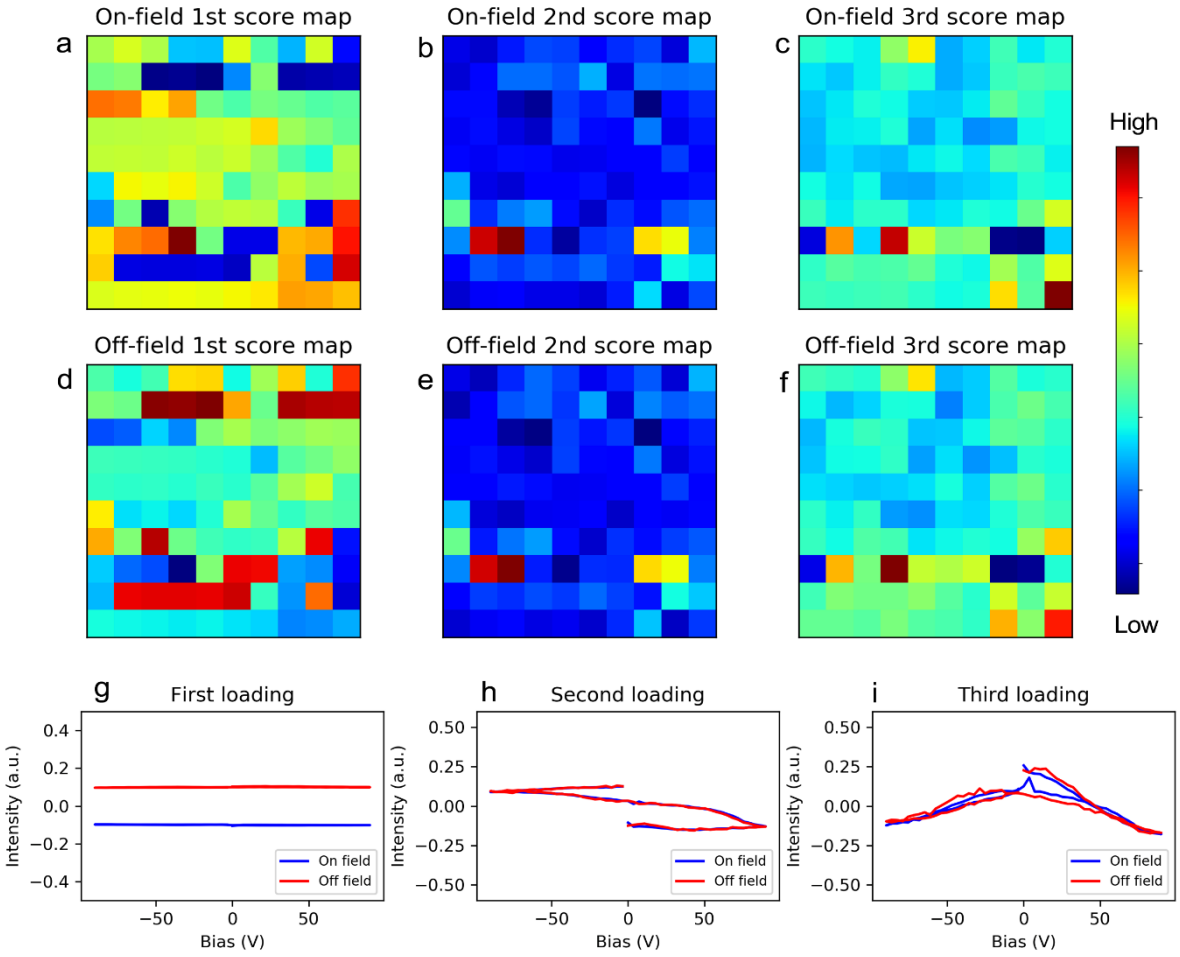

Fig. S6 Principal component analysis (PCA) on the contact resonance frequency (f_0) of glass in the SM-ESM measurements: (a-c) The score maps for the first three principal components at the on-field; (d-f) The score maps for the first three principal components at the off-field; and (g-i) The loadings for the first three principal components. The percentages of variance explained by the first, the second, and the third on-field principal components are 99.68%, 0.22%, and 0.05%, respectively; at the off field, they are 99.64%, 0.22%, and 0.06%, respectively. The SM-ESM measurement is scanned in a 10 × 10 μm^2 area with a resolution of 10 × 10 points.

Table S1. Pearson's correlation coefficients (r) of the PCA score maps, and the cosine similarity (s) of the PCA loadings of the ferroelectric materials. The PCA score maps and loadings are decomposed from the SS-PFM contact resonance frequency (f_0) dataset obtained from PZN-4.5%PT, PZN-7%PT, PZN-9%PT, and BLGF-35%PT. All the scanned areas are with spontaneous polarization.

		r of score maps			s of loadings		
	On \ Off	1st	2nd	3rd	1st	2nd	3rd
PZN-4.5%PT	1st	1.00**	0.01	-0.03	1.00	0.00	-0.01
	2nd	0.04	-0.84**	0.2**	0.01	-0.76	-0.16
	3rd	-0.04	-0.47**	-0.41**	0.02	-0.36	-0.06
PZN-7%PT	1st	1.00**	0.06	-0.02	1.00	-0.05	-0.01
	2nd	0.05	-0.04	0.48**	0.03	0.35	0.43
	3rd	-0.04	0.77**	0.20**	-0.01	0.28	-0.51
PZN-9%PT	1st	1.00**	0.01	0.01	0.97	-0.02	-0.07
	2nd	-0.00	-0.28**	0.38**	-0.03	0.01	0.05
	3rd	0.00	0.55**	-0.44**	0.00	-0.01	-0.01
BLGF-35%PT	1st	0.96**	0.17**	-0.13**	0.98	0.04	0.09
	2nd	-0.25**	0.58**	-0.49**	-0.09	-0.13	0.13
	3rd	-0.00	0.69**	0.64**	0.06	0.57	0.21

p-value of the Pearson's correlation: (*) for $p < 0.05$, (**) for $p < 0.01$

Table S2. Pearson's correlation coefficients (r) of the first, second and third PCA score maps, and the cosine similarity (s) of the first three PCA loadings of the non-ferroelectric materials. The PCA score maps and loadings are decomposed from the contact resonance frequency (f_0) dataset of the SM-ESM measurements obtained from glass and TiO_2.

		r of score maps			s of loadings		
	On \ Off	1st	2nd	3rd	1st	2nd	3rd
Glass	1st	-1.00**	0.00	0.00	-1.00	0.00	0.01
	2nd	0.00	1.00**	0.01	0.00	1.00	-0.02
	3rd	0.00	-0.01	0.97**	0.00	0.01	0.97
TiO_2	1st	0.99**	0.02	-0.02	1.00	-0.01	0.00
	2nd	0.03	-0.94**	0.05	-0.01	-1.00	0.04
	3rd	-0.00	0.12	-0.79**	0.00	-0.03	-0.97

p-value of the Pearson's correlation: (*) for $p < 0.05$, (**) for $p < 0.01$

Table S3. The percentages of variance explained by the first, second and third principal components of the ferroelectric materials.

Sample	Field	1st	2nd	3rd
PZN-4.5%PT (poled by DC)	On	83.58%	4.70%	3.87%
	Off	90.80%	2.88%	1.27%
PZN-4.5%PT (spontaneous polarization)	On	87.95%	3.74%	1.20%
	Off	91.35%	2.87%	0.85%
PZN-7%PT	On	92.73%	1.47%	1.11%
	Off	90.51%	1.91%	1.56%
PZN-9%PT	On	83.93%	5.33%	4.09%
	Off	88.08%	2.48%	1.94%
BLGF-35%PT	On	68.04%	12.38%	6.31%
	Off	76.17%	9.96%	4.59%

Table S4. The percentages of variance explained by the first, second and third principal components of the non-ferroelectric materials.

Sample	Field	1st	2nd	3rd
PMMA	On	98.88%	0.42%	0.21%
	Off	98.98%	0.43%	0.20%
glass	On	99.68%	0.22%	0.05%
	Off	99.64%	0.22%	0.06%
Al_2O_3	On	95.40%	2.92%	0.93%
	Off	95.60%	2.82%	0.79%
TiO_2	On	93.78%	4.70%	1.01%
	Off	94.24%	4.50%	0.74%