

## Supporting Information

### Supercritical relaxor phase boundary for ultrahigh electrostrictive properties

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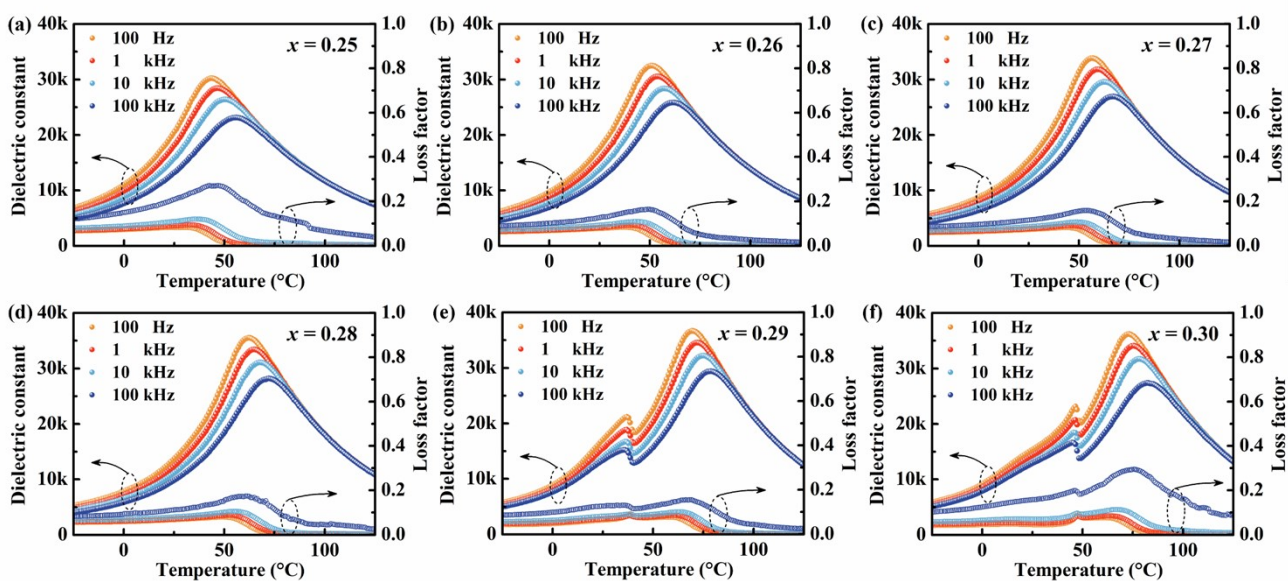


Fig. S1 The temperature and frequency dependence of dielectric permittivity and dielectric loss for 3%Nd-PMN-xPT ceramics: (a)  $x = 0.25$ , (b)  $x = 0.26$ , (c)  $x = 0.27$ , (d)  $x = 0.28$ , (e)  $x = 0.29$ , (f)  $x = 0.30$ .

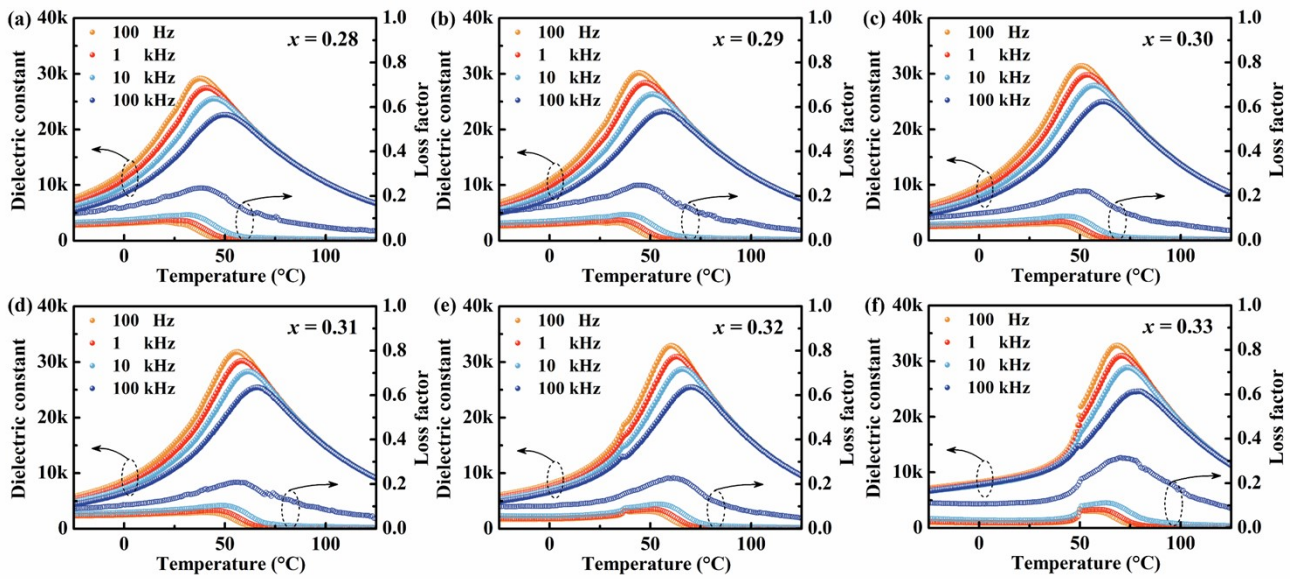


Fig. S2 The temperature and frequency dependence of dielectric permittivity and dielectric loss for 4%Nd-PMN- $x$ PT ceramics: (a)  $x = 0.28$ , (b)  $x = 0.29$ , (c)  $x = 0.30$ , (d)  $x = 0.31$ , (e)  $x = 0.32$ , (f)  $x = 0.33$ .

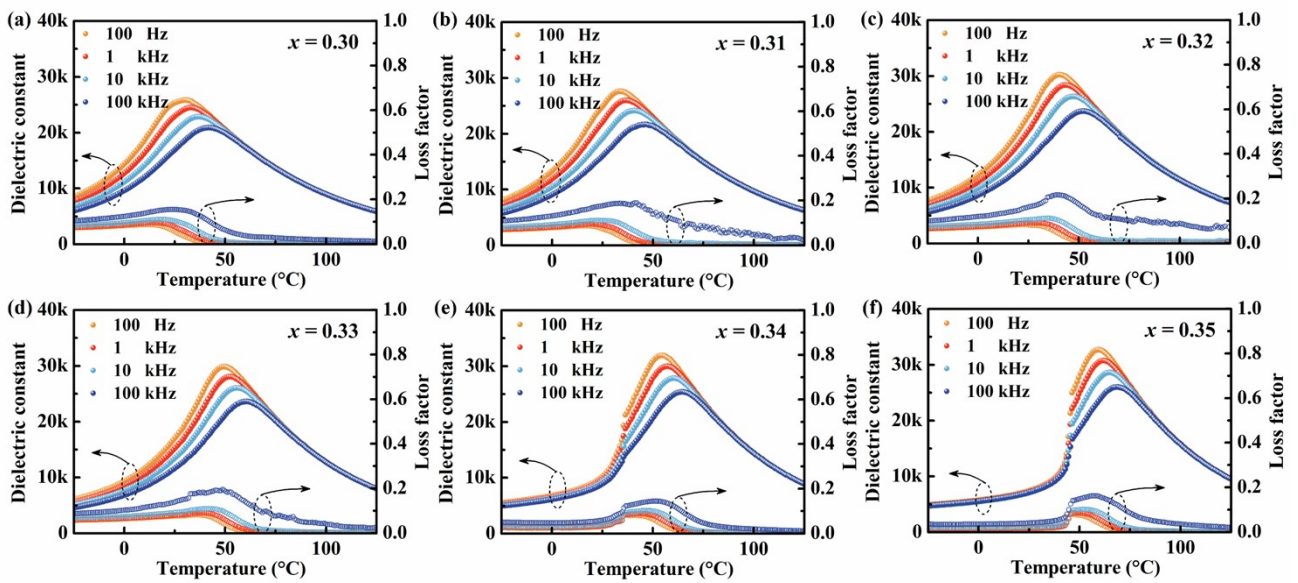


Fig. S3 The temperature and frequency dependence of dielectric permittivity and dielectric loss for 5%Nd-PMN- $x$ PT ceramics: (a)  $x = 0.30$ , (b)  $x = 0.31$ , (c)  $x = 0.32$ , (d)  $x = 0.33$ , (e)  $x = 0.34$ , (f)  $x = 0.35$ .

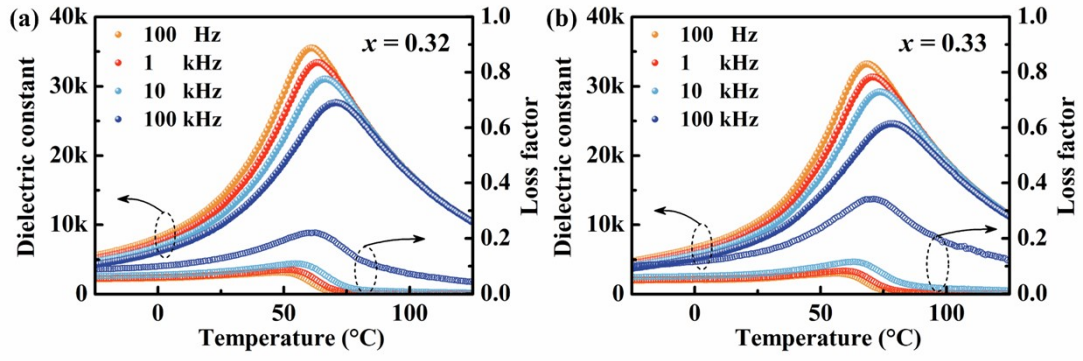


Fig. S4 The temperature dependence of dielectric permittivity and dielectric loss for unpoled (a) 4%Nd-PMN-0.32PT and (b) 4%Nd-PMN-0.33PT ceramic.

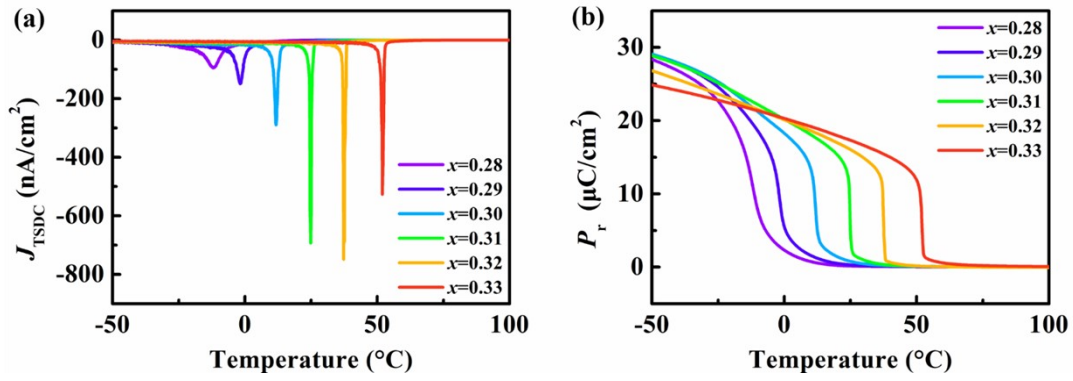


Fig. S5 The temperature stimulated depolarization current density  $J_{TSDC}$  and calculated remanent polarization  $P_r$  for 4%Nd-PMN- $x$ PT ceramics.

The remanent polarization  $P_r$  was calculated by the formula:

$$P_r = \int_0^T \frac{J}{r} dT$$

where  $J$ ,  $r$ ,  $T$  are the depolarization current density, heating rate, and temperature, respectively.

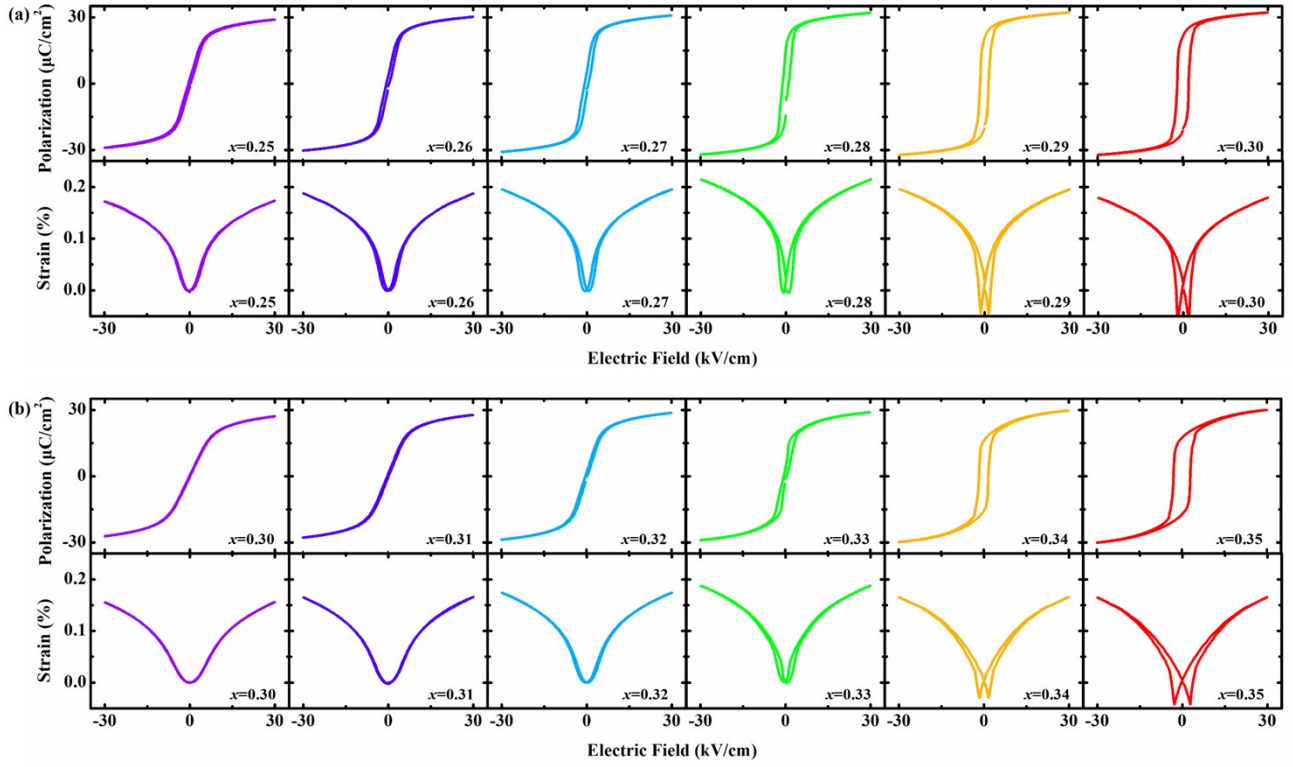


Fig. S6 The polarization-electric field ( $P$ - $E$ ) hysteresis loops and the bipolar strain-electric field ( $S$ - $E$ ) curves for (a) 3%Nd-PMN- $x$ PT ceramics and (b) 5%Nd-PMN- $x$ PT ceramics.

Table S1 The dielectric, piezoelectric and strain properties of yNd-PMN-xPT ferroelectric ceramics

Composition	$\epsilon_r$ (1kHz)	$\tan \delta$ (%)	$T_C$ (°C)	$T_{nr}$ (°C)	$d_{33}$ (pC/N)	$\Delta T_m$ (°C)	$S_{max}$ (%)
3%Nd-PMN-0.25PT	18230	9.0	46	-	-	12.1	0.178
3%Nd-PMN-0.26PT	15800	8.4	52	-	-	11.0	0.189
3%Nd-PMN-0.27PT	14020	7.6	58	-	-	10.6	0.195
3%Nd-PMN-0.28PT	12520	6.9	64	-	-	10.5	0.210
3%Nd-PMN-0.29PT	15510	6.7	71	37	820	9.3	0.195
3%Nd-PMN-0.30PT	14330	5.3	76	46	1270	8.9	0.179
4%Nd-PMN-0.28PT	20620	9.3	40	-	-	12.3	0.186
4%Nd-PMN-0.29PT	17630	8.7	46	-	-	11.6	0.194
4%Nd-PMN-0.30PT	15920	7.7	53	-	-	10.8	0.201
4%Nd-PMN-0.31PT	14050	7.2	58	-	-	10.1	0.217
4%Nd-PMN-0.32PT	11750	5.4	63	37	430	9.7	0.198
4%Nd-PMN-0.33PT	9740	2.3	70	50	890	9.2	0.188
5%Nd-PMN-0.30PT	23050	7.7	32	-	-	12.5	0.155
5%Nd-PMN-0.31PT	22370	8.8	36	-	-	11.5	0.165
5%Nd-PMN-0.32PT	20320	9.0	43	-	-	11.3	0.173
5%Nd-PMN-0.33PT	16430	8.0	51	-	-	10.8	0.187
5%Nd-PMN-0.34PT	9930	3.4	56	36	220	10.3	0.168
5%Nd-PMN-0.35PT	7350	1.8	61	46	450	9.5	0.163