Electronic supplementary information (ESI)

Strain-induced giant enhancement of anisotropic dielectric constant in layered nitrides $SrHfN_2$ and $SrZrN_2$

Min Yao,[1,2] Yuanyun Zhang,[1] Jianmin Ban,[1] Junjie Hou,[1] Bowen Zhang,[3] Junwei Liu,[3] Xing Ming,[1,2,3*] Xiaojun Kuang[3†]

1. *College of Science, Guilin University of Technology, Guilin 541004, People's Republic of China*
2. *Key Laboratory of Low-dimensional Structural Physics and Application, Education Department of Guangxi Zhuang Autonomous Region, Guilin University of Technology, Guilin 541004, People's Republic of China*
3. *Key Lab of New Processing Technology for Nonferrous Metal & Materials, Ministry of Education, School of Materials Science and Engineering, Guilin University of Technology, Guilin 541004, China.*

[*] **Email:** kuangxj@glut.edu.cn

[†] **Email:** kuangxj@glut.edu.cn

Table S1. The evolutions of Sr-N, Hf-N and Zr-N bond lengths *d* (Å) and octahedral distortion parameters Σ (°) and Θ (°) of the SrN_6, HfN_6 and ZrN_6 octahedra in $SrHfN_2$ and $SrZrN_2$ under isotropic strain.

Strain (%)	$SrHfN_2$						$SrZrN_2$					
	SrN_6			HfN_6			SrN_6			ZrN_6		
	d	Σ	Θ	*d*	Σ	Θ	*d*	Σ	Θ	*d*	Σ	Θ
-1.0	2.600	130.685	269.201	2.234	67.856	168.050	2.601	123.073	255.060	2.255	64.740	159.789
-0.9	2.603	130.924	269.642	2.236	68.138	168.801	2.604	123.303	255.488	2.257	65.017	160.519
-0.8	2.606	131.164	270.084	2.238	68.423	169.558	2.607	123.539	255.930	2.258	65.285	161.228
-0.7	2.610	131.405	270.530	2.240	68.710	170.325	2.611	123.777	256.373	2.260	65.574	161.993
-0.6	2.613	131.648	270.978	2.241	69.001	171.101	2.614	124.013	256.814	2.262	65.847	162.716
-0.5	2.616	131.892	271.427	2.243	69.288	171.867	2.617	124.249	257.255	2.264	66.130	163.466
-0.4	2.619	132.136	271.878	2.245	69.580	172.645	2.620	124.487	257.699	2.266	66.403	164.191
-0.3	2.622	132.381	272.329	2.247	69.871	173.424	2.623	124.724	258.140	2.268	66.688	164.947
-0.2	2.626	132.628	272.783	2.249	70.165	174.209	2.627	124.966	258.591	2.269	66.966	165.684
-0.1	2.629	132.875	273.238	2.251	70.459	174.996	2.630	125.204	259.034	2.271	67.254	166.449
0	2.632	133.128	273.704	2.252	70.762	175.808	2.633	125.445	259.484	2.273	67.538	167.203
0.1	2.635	133.376	274.161	2.254	71.057	176.598	2.636	125.688	259.935	2.275	67.825	167.968

0.2	2.639	133.729	274.810	2.256	71.477	177.723	2.639	125.932	260.390	2.277	68.104	168.711
0.3	2.642	133.986	275.283	2.258	71.785	178.550	2.642	126.176	260.844	2.279	68.401	169.502
0.4	2.645	134.245	275.758	2.259	72.093	179.377	2.646	126.424	261.304	2.281	68.685	170.259
0.5	2.648	134.505	276.235	2.261	72.404	180.213	2.649	126.669	261.759	2.282	68.983	171.052
0.6	2.652	134.765	276.713	2.263	72.716	181.050	2.652	126.919	262.224	2.284	69.273	171.827
0.7	2.655	135.027	277.194	2.265	73.029	181.893	2.655	127.165	262.682	2.286	69.570	172.620
0.8	2.658	135.297	277.689	2.267	73.352	182.763	2.659	127.416	263.147	2.288	69.859	173.392
0.9	2.661	135.450	277.970	2.268	73.534	183.255	2.662	127.666	263.611	2.290	70.162	174.201
1	2.665	135.830	278.667	2.270	73.990	184.483	2.665	127.919	264.080	2.292	70.454	174.983
1.1	2.668	136.081	279.127	2.272	74.291	185.296	2.668	128.171	264.548	2.293	70.753	175.783
1.2	2.671	136.346	279.613	2.274	74.609	186.154	2.671	128.425	265.019	2.295	71.054	176.589
1.3	2.674	136.590	280.058	2.275	74.901	186.943	2.675	128.680	265.491	2.297	71.356	177.399
1.4	2.678	136.853	280.540	2.277	75.216	187.796	2.678	128.936	265.966	2.299	71.660	178.215
1.5	2.681	137.117	281.023	2.279	75.533	188.654	2.681	129.194	266.443	2.301	71.965	179.034
1.6	2.684	137.382	281.508	2.281	75.851	189.515	2.684	129.453	266.922	2.302	72.273	179.860

Table S2. Calculated diagonal components of the dielectric tensors and dielectric constants of $SrHfN_2$. The crystal structure becomes unstable when isotropic strain exceeds 1.2%, which results in unreasonable dielectric constants.

Strain(%)	ε_{ele}			ε_{ion}			ε_r
	x	y	z	x	y	z	
-1.0	12.36	12.36	11.93	125.70	125.70	46.70	111.59
-0.9	12.29	12.29	11.85	130.72	130.72	47.38	115.08
-0.8	12.22	12.22	11.77	136.27	136.27	48.09	118.94
-0.7	12.15	12.15	12.15	142.42	142.41	48.83	123.37
-0.6	12.08	12.08	11.62	149.21	149.21	49.63	127.95
-0.5	12.02	12.02	11.55	156.80	156.80	50.47	133.22
-0.4	11.96	11.96	11.48	165.36	165.35	51.35	139.15
-0.3	11.90	11.90	11.41	175.08	175.07	52.29	145.88
-0.2	11.84	11.84	11.35	186.16	186.16	53.28	153.55
-0.1	11.79	11.79	11.29	198.82	198.80	54.33	162.27
0	11.89	11.89	11.30	217.77	217.78	56.02	175.55
0.1	11.84	11.84	11.24	235.50	235.51	57.20	187.71
0.2	11.78	11.78	11.19	253.88	253.87	58.55	200.35
0.3	11.73	11.73	11.14	278.65	278.66	59.88	217.27
0.4	11.68	11.68	11.08	309.15	309.13	61.30	238.01
0.5	11.63	11.63	11.03	347.65	347.61	62.82	264.13
0.6	11.59	11.59	10.99	398.01	397.95	64.45	298.19
0.7	11.54	11.54	10.94	466.05	466.05	66.17	344.10
0.8	11.50	11.50	10.90	562.14	562.04	68.02	408.70
0.9	11.46	11.46	10.85	735.64	735.64	69.82	524.95
1	11.42	11.42	10.82	969.30	968.94	72.09	681.33
1.1	11.38	11.38	10.78	1553.61	1554.54	74.32	1072.00
1.2	11.34	11.34	10.74	3889.58	3878.54	76.76	2626.10
1.3	11.30	11.30	10.70	1.98	1.98	79.33	38.87
1.4	11.27	11.27	10.67	2.10	2.10	82.14	39.84
1.5	11.23	11.23	10.63	2.20	2.20	85.17	40.89
1.6	11.20	11.20	10.60	2.31	2.31	88.50	42.04

Table S3. Calculated diagonal components of the dielectric tensors and dielectric constants of $SrZrN_2$. The crystal structure becomes unstable when isotropic strain exceeds 0.7%, which results in unreasonable dielectric constants.

Strain(%)	ε_{ele}			ε_{ion}			ε_r
	x	y	z	x	y	z	
-1.0	13.96	13.96	12.92	183.98	183.97	50.85	153.21
-0.9	13.85	13.85	12.80	193.45	193.45	51.54	159.65
-0.8	13.75	13.75	12.70	204.09	203.87	52.27	166.81
-0.7	13.92	13.92	12.73	222.67	222.64	53.87	179.92
-0.6	13.82	13.82	12.63	236.96	237.03	54.69	189.65
-0.5	13.72	13.72	12.53	253.72	253.69	55.57	200.98
-0.4	13.63	13.63	12.43	273.29	273.28	56.49	214.25
-0.3	13.53	13.53	12.34	297.00	297.11	57.47	230.33
-0.2	13.45	13.45	12.25	325.24	324.16	58.51	249.02
-0.1	13.37	13.37	12.17	360.05	359.08	59.61	272.55
0	13.29	13.29	12.09	403.74	403.74	60.79	302.31
0.1	13.21	13.21	12.01	461.39	460.57	62.04	340.81
0.2	13.13	13.13	11.94	537.80	539.75	63.36	393.04
0.3	13.06	13.06	11.87	647.85	647.66	64.78	466.10
0.4	12.99	12.99	11.80	817.28	814.73	66.29	578.70
0.5	12.93	12.93	11.73	1104.54	1104.42	67.89	771.48
0.6	12.87	12.87	11.67	1723.03	1699.23	69.60	1176.42
0.7	12.80	12.80	11.61	4019.66	4017.37	71.44	2715.23
0.8	12.74	12.74	11.55	1.17	1.17	73.40	37.60
0.9	12.69	12.69	11.50	1.26	1.26	75.53	38.31
1	12.63	12.63	11.44	1.35	1.35	77.77	39.06
1.1	12.58	12.58	11.39	1.44	1.43	80.21	39.88
1.2	12.53	12.53	11.34	1.52	1.52	82.81	40.75
1.3	12.48	12.48	11.30	1.61	1.61	85.70	41.73
1.4	12.43	12.43	11.25	1.69	1.69	88.76	42.75
1.5	12.38	12.38	11.21	1.79	1.79	92.16	43.91
1.6	12.34	12.34	11.16	1.87	1.87	95.80	45.13

Table S4. Calculated zone-center phonon frequencies of the three acoustic branches, which including one longitudinal and two transverse acoustic branches (LA, TA1 and TA2), the TA branches show obviously imaginary frequencies when the isotropic strain exceeds 1.2% (0.7%) for $SrHfN_2$ ($SrZrN_2$), indicating the lattice structures of $SrHfN_2$ and $SrZrN_2$ become unstable.

Strain(%)	$SrHfN_2$			$SrZrN_2$		
	LA(cm^{-1})	TA1(cm^{-1})	TA2(cm^{-1})	LA(cm^{-1})	TA1(cm^{-1})	TA2(cm^{-1})
0.8				0.05	13.85	13.85
0.9				0.05	30.87	30.87
1				0.05	41.46	41.46
1.1				0.05	49.77	49.77
1.2				0.03	56.92	56.92
1.3	0.02	14.59	14.59	0.05	63.46	63.46
1.4	0.03	28.59	28.60	0.01	69.22	69.22
1.5	0.02	37.76	37.77	0.01	74.70	74.70
1.6	0.02	45.35	45.36	0.02	79.69	79.69

Table S5. Mode frequencies ω_λ (cm^{-1}) and effective charges $\widetilde{Z}^*_\lambda$ (in |e|) of the IR-active modes for $SrHfN_2$ and $SrZrN_2$ under isotropic strain. The appearance of imaginary frequencies of the two TA branches is accompanied by the zero-frequency $E_u(2)$ mode when the strain increases to 1.3% (0.8%) for $SrHfN_2$ and $SrZrN_2$.

Strain	$SrHfN_2$								$SrZrN_2$							
	A2u(1)		A2u(2)		Eu(1)		Eu(2)		A2u(1)		A2u(2)		Eu(1)		Eu(2)	
	ω_λ	$\widetilde{Z}^*_\lambda$	ω_λ	$\widetilde{Z}^*_\lambda$	ω_λ	$\widetilde{Z}^*_\lambda$	ω_λ	$\widetilde{Z}^*_\lambda$	ω_λ	$\widetilde{Z}^*_\lambda$	ω_λ	$\widetilde{Z}^*_\lambda$	ω_λ	$\widetilde{Z}^*_\lambda$	ω_λ	$\widetilde{Z}^*_\lambda$
-1	322.78	1.31	190.89	1.46	175.55	3.08	99.33	0.41	352.98	1.09	194.07	1.86	161.34	2.81	108.87	1.21
-0.9	321.69	1.30	188.70	1.46	173.16	3.03	97.74	0.45	351.92	1.07	191.82	1.86	159.33	2.69	106.27	1.31
-0.8	320.61	1.28	186.51	1.46	170.78	2.98	96.07	0.48	350.93	1.06	189.48	1.85	157.42	2.58	103.78	1.40
-0.7	319.54	1.26	184.30	1.47	168.39	2.93	94.34	0.52	349.72	1.05	186.56	1.86	154.90	2.46	100.43	1.56
-0.6	318.47	1.24	182.06	1.47	166.01	2.88	92.53	0.56	348.72	1.04	184.19	1.86	153.11	2.35	97.53	1.66
-0.5	317.40	1.22	179.80	1.47	163.64	2.82	90.64	0.61	347.70	1.03	181.85	1.85	151.33	2.23	94.31	1.76
-0.4	316.34	1.21	177.54	1.48	161.29	2.76	88.64	0.66	346.81	1.02	179.28	1.85	149.64	2.13	91.61	1.83
-0.3	315.28	1.19	175.23	1.48	158.94	2.69	86.53	0.71	345.69	1.00	177.08	1.84	147.91	2.00	87.34	1.95
-0.2	314.22	1.17	172.91	1.48	156.60	2.62	84.30	0.77	344.75	0.99	174.58	1.84	146.28	1.90	83.95	2.04
-0.1	313.18	1.16	170.56	1.48	154.31	2.56	81.95	0.83	343.69	0.98	172.24	1.83	144.67	1.79	79.48	2.14

0	311.99	1.14	167.78	1.50	151.61	2.50	79.23	0.91	342.66	0.97	169.87	1.83	143.13	1.68	74.80	2.23
0.1	310.96	1.13	165.39	1.50	149.38	2.42	76.53	0.97	341.71	0.96	167.31	1.82	141.62	1.59	70.25	2.30
0.2	310.01	1.11	162.82	1.50	147.26	2.35	73.98	1.03	340.64	0.95	164.96	1.82	140.18	1.49	64.34	2.39
0.3	308.99	1.10	160.39	1.51	145.10	2.27	70.93	1.10	339.73	0.94	162.28	1.81	138.73	1.41	59.23	2.45
0.4	307.98	1.08	157.93	1.51	142.98	2.19	67.64	1.18	338.74	0.93	159.75	1.81	137.37	1.33	52.70	2.52
0.5	306.97	1.07	155.42	1.51	140.89	2.10	64.07	1.25	337.76	0.92	157.15	1.81	136.03	1.26	45.32	2.58
0.6	305.96	1.05	152.87	1.52	138.86	2.02	60.13	1.33	336.75	0.91	154.61	1.80	134.74	1.18	35.90	2.64
0.7	304.96	1.04	150.31	1.52	136.85	1.93	55.80	1.40	335.80	0.91	151.90	1.80	133.44	1.12	23.73	2.69
0.8	303.98	1.03	147.72	1.52	134.93	1.84	51.02	1.48	334.82	0.90	149.24	1.79	132.21	1.06	0.08	0.00
0.9	302.93	1.01	145.29	1.52	133.01	1.75	44.79	1.56	333.84	0.89	146.53	1.79	130.99	1.00	0.00	0.00
1	302.03	1.00	142.46	1.53	131.21	1.68	39.17	1.63	332.87	0.88	143.81	1.79	129.80	0.94	0.01	0.00
1.1	301.05	0.99	139.82	1.53	129.42	1.59	31.09	1.71	331.91	0.87	141.05	1.79	128.65	0.89	0.01	0.00
1.2	300.08	0.98	137.09	1.53	127.67	1.51	19.71	1.78	330.96	0.86	138.25	1.78	127.53	0.85	0.09	0.00
1.3	299.11	0.97	134.40	1.53	125.98	1.43	0.10	0.00	329.98	0.85	135.39	1.78	126.40	0.80	0.01	0.00
1.4	298.16	0.95	131.62	1.54	124.34	1.35	0.01	0.00	329.03	0.85	132.48	1.78	125.31	0.76	0.10	0.00
1.5	297.22	0.94	128.82	1.54	122.76	1.28	0.02	0.00	328.06	0.84	129.53	1.78	124.23	0.73	0.00	0.00
1.6	296.27	0.93	125.95	1.54	121.20	1.21	0.03	0.00	327.12	0.83	126.54	1.77	123.19	0.69	0.02	0.00

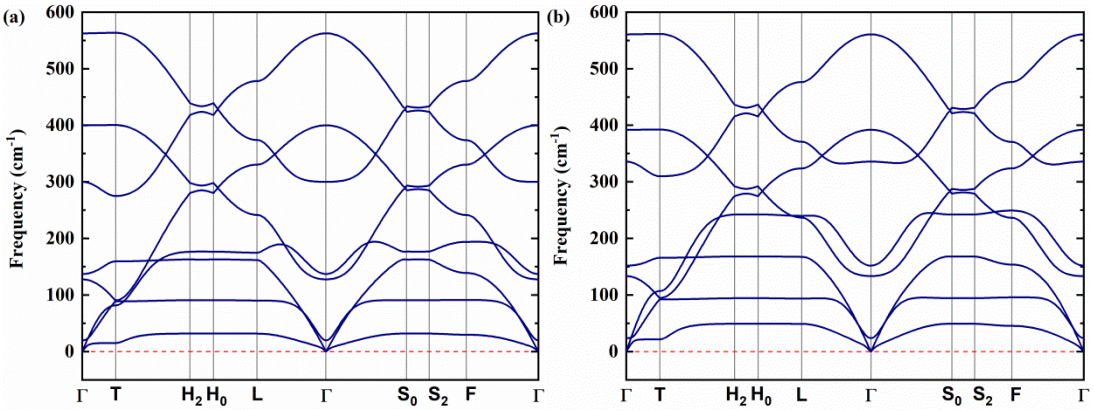

Figure S1. The phonon spectra of (a) $SrHfN_2$ and (b) $SrZrN_2$ under isotropic strain of 1.2% and 0.7%.

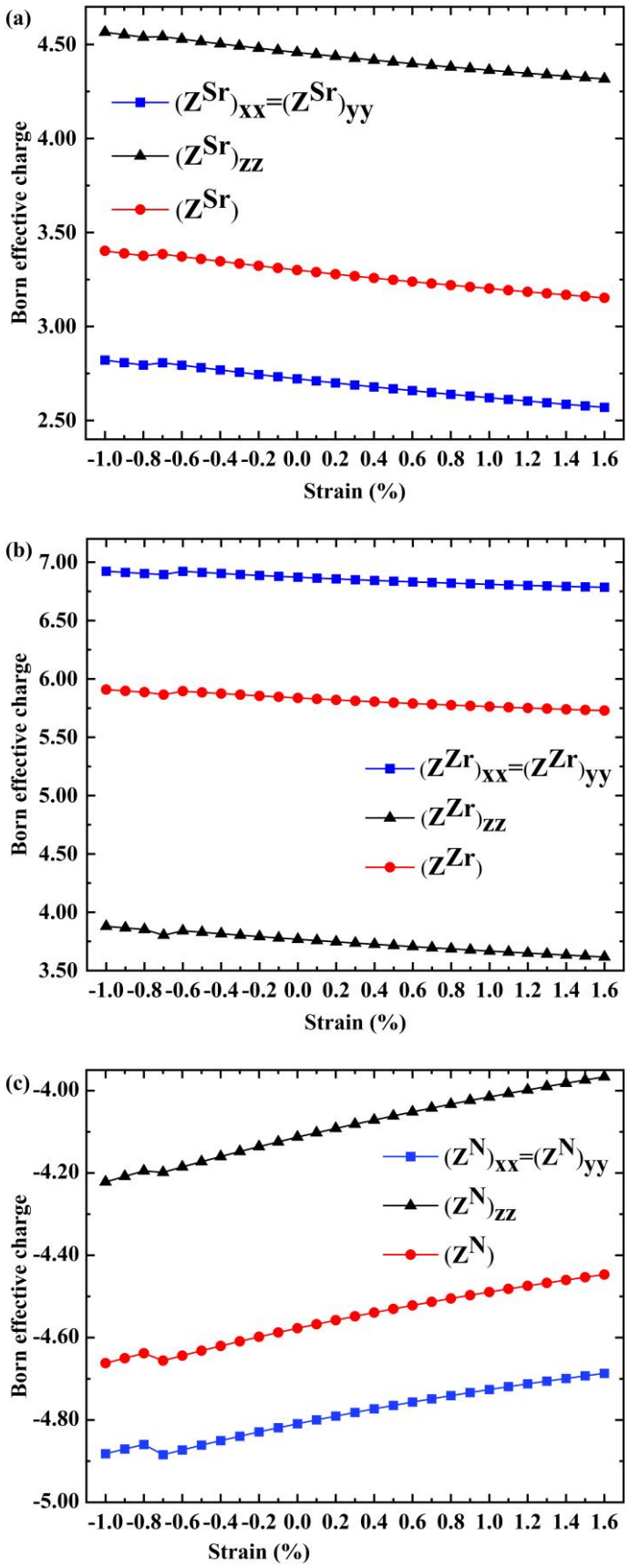

Figure S2. Born effective charges components of each element and its average value under isotropic strain for $SrZrN_2$: (a) Sr, (b) Zr and (c) N.

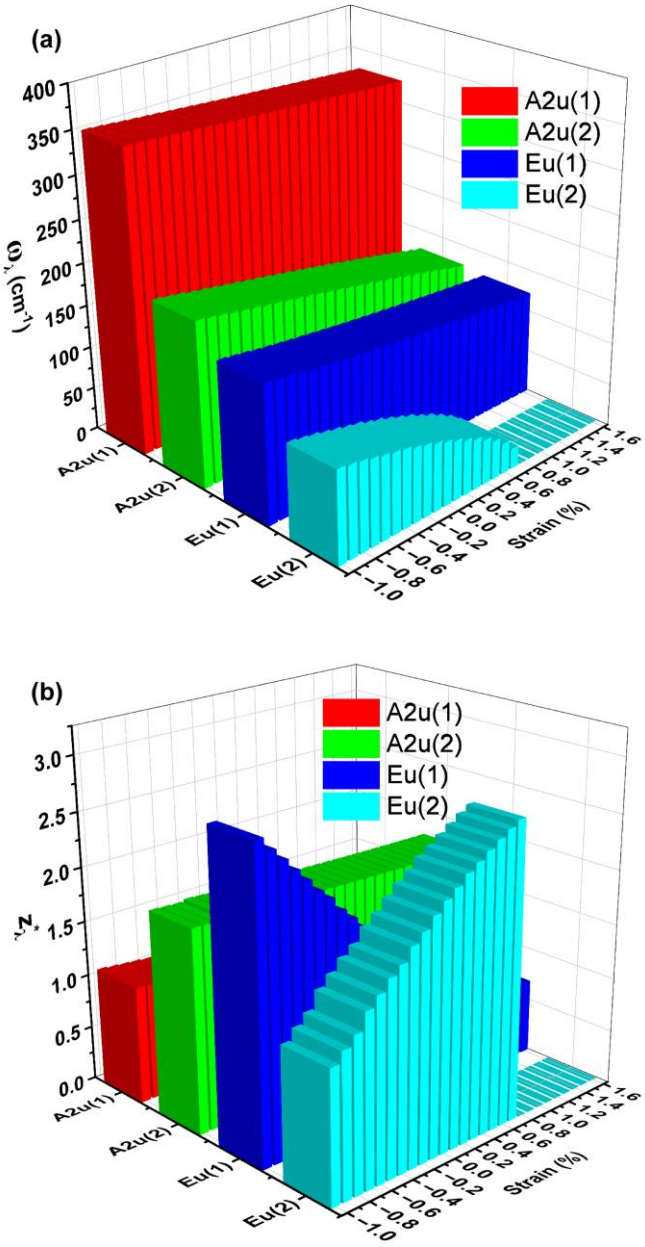

Figure S3. (a) Mode frequencies ω_λ and (b) effective charges $\widetilde{Z_\lambda^*}$ (in |e|) of the IR-active modes for $SrZrN_2$ under isotropic strain. The IR activity frequency of the $E_u(2)$ mode reduces by about 2/3 from 74.7 to 23.7 when the tensile strain increases up to 0.7%, and the corresponding mode effective charge increases by about 20%, leading to a giant dielectric constant of 2700 under 0.7% tensile strain.