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Supporting Information

# Ultra-thin lead oxide piezoelectric layers for reduced environmental contamination using a liquid metal-based process

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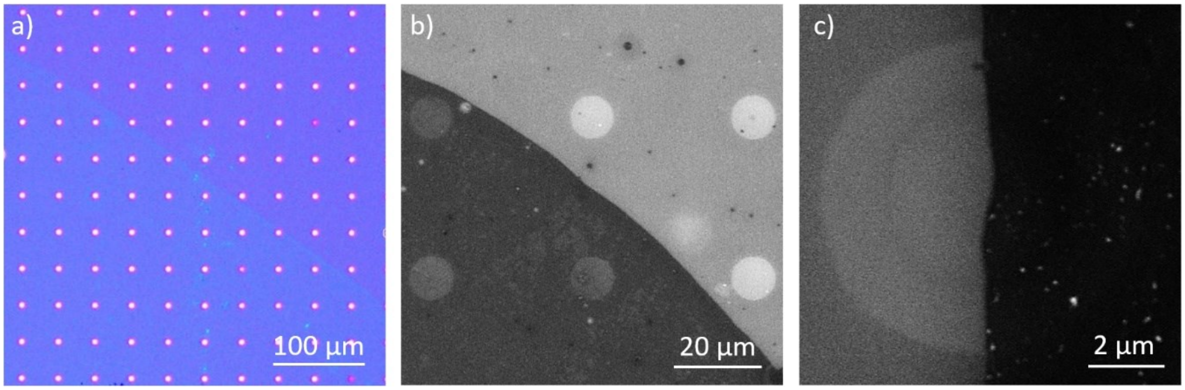


**Fig. S1** (a) Optical and (b,c) SEM images of the 2D PbO on a holey $SiO_2$/Si substrate.

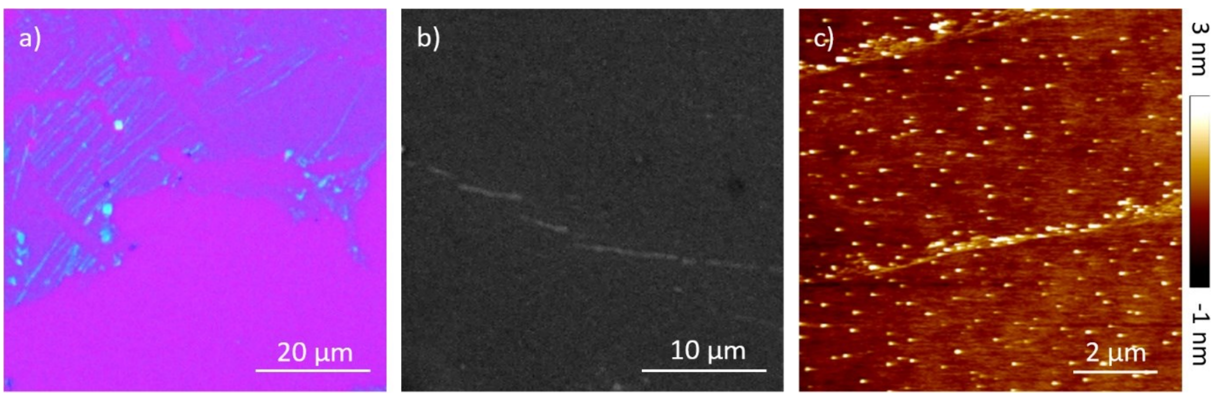


**Fig. S2** (a) Optical, (b) SEM and (c) AFM images of the 2D PbO wrinkles on a $SiO_2$/Si substrate.

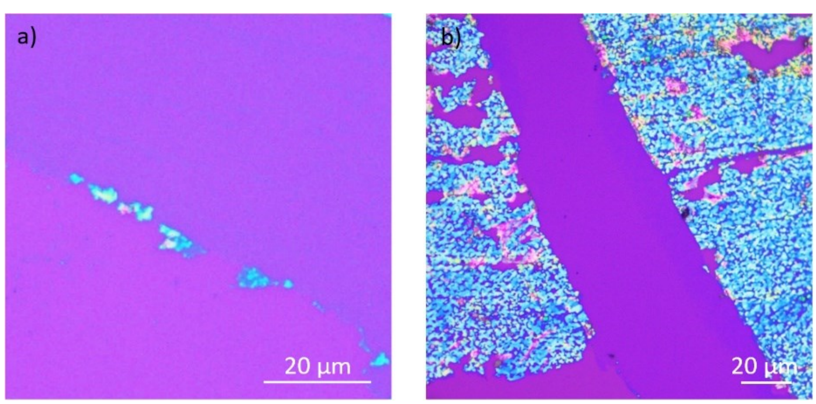


**Fig. S3** Bulk lead oxides on the edges of monolayer PbO after (a) short and (b) long (15 min) oxidation periods.

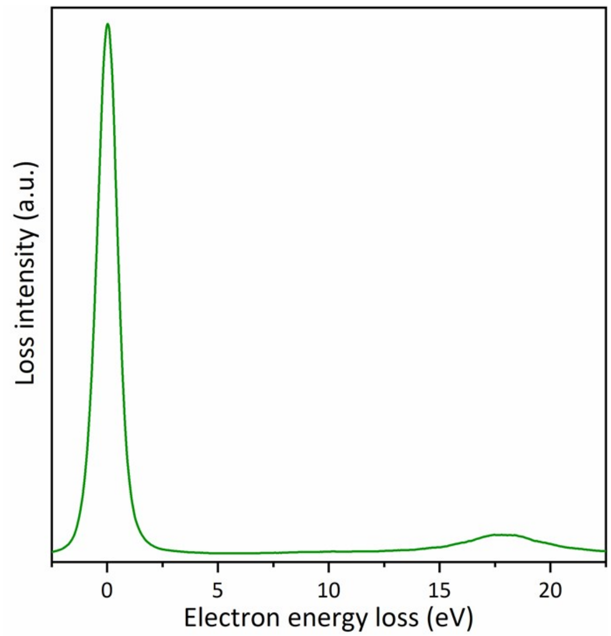


**Fig. S4** The EELS spectrum of 2D PbO nanosheets.

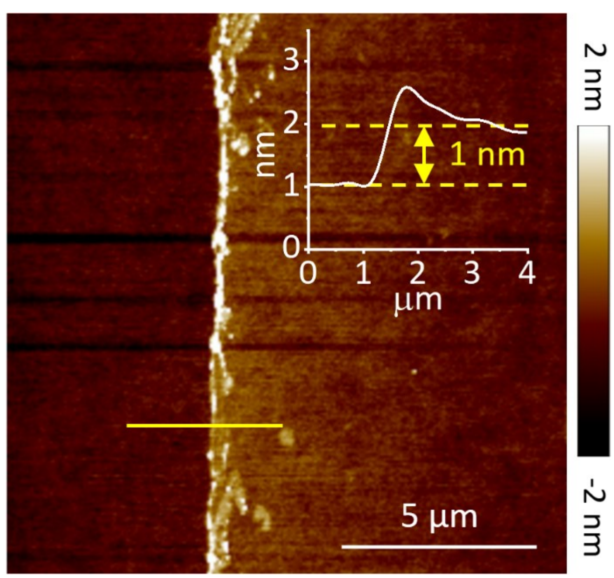


**Fig. S5** The AFM image of the area of 2D PbO used for piezoelectricity measurements.

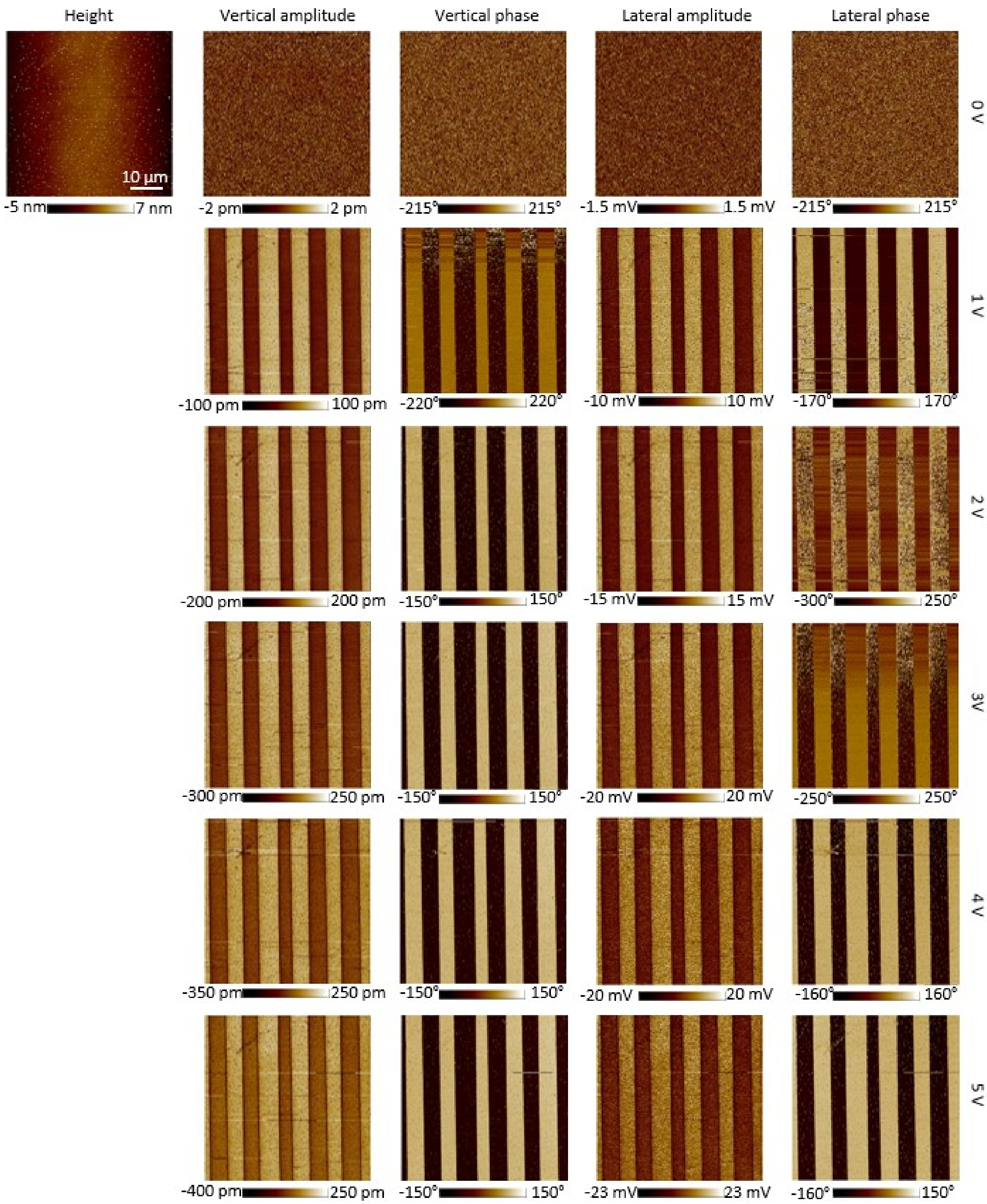


**Fig. S6** Topography, amplitude, and phase variation of PPLN as the function of magnitudes of AC driving voltages.

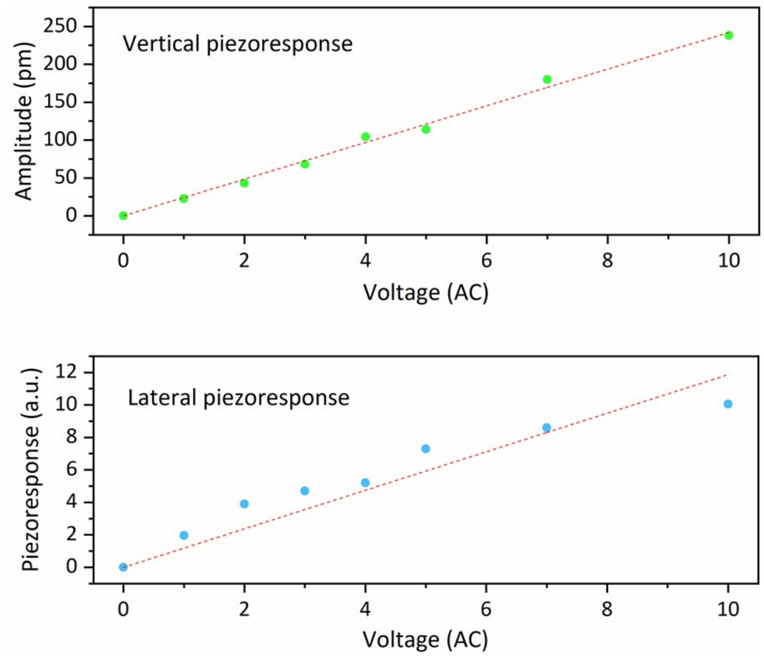


**Fig. S7** Extended vertical and lateral piezoresponse amplitudes of the PbO monolayer as the function of AC driving voltage.

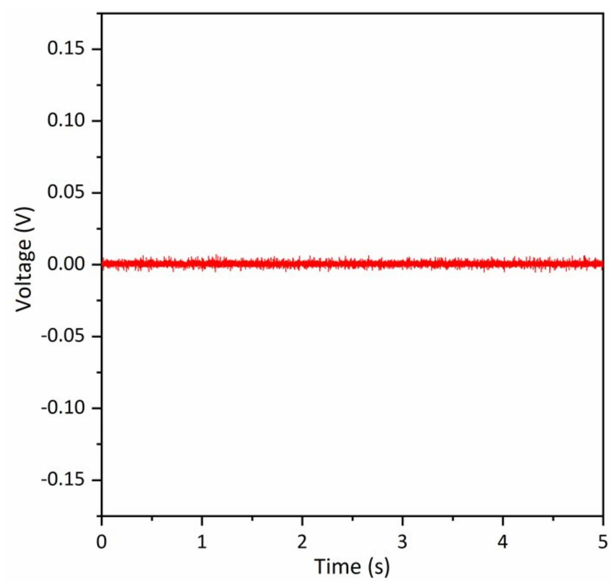


**Fig. S8** Voltage output from the bare (no PbO layer) mica device after applying mechanical stimuli.

**Table S1.** Computed piezoelectric stress tensor (in C $m^{-2}$) and relevant elastic tensors (in GPa) components for a metastable β-PbO structure.

| $e_{31}$ | $e_{32}$ | $e_{33}$ | $C_{11}$ | $C_{12}$ | $C_{13}$ | $C_{22}$ | $C_{23}$ | $C_{33}$ |
|---|---|---|---|---|---|---|---|---|
| 0.069 | 0.069 | -0.173 | 4.4 | 0.8 | 0.8 | 11.8 | 3.0 | 7.4 |

**Table S2.** Vertical and lateral piezoresponses of some 2D materials.

| Material | Vertical piezoresponse (pm/V) | Lateral piezoresponse (pm/V) | Ref. | Material | Vertical piezoresponse (pm/V) | Lateral piezoresponse (pm/V) | Ref. |
|---|---|---|---|---|---|---|---|
| $C_3N_4$ | 1 | | 1 | GaSe | 2.30 | | 2 |
| MoSSe | | 0.020 | 3 | GaS | 2.06 | | 2 |
| MoSTe | | 0.028 | 3 | InSe | 1.46 | | 2 |
| MoSeTe | | 0.030 | 3 | Graphene oxide | | 0.24 | 4 |
| WSSe | | 0.011 | 3 | $GaInTe_2$ | | 0.32 | 5 |
| WSTe | | 0.007 | 3 | $GaInSe_2$ | | 0.46 | 5 |
| WSeTe | | 0.008 | 3 | $GaInS_2$ | | 0.38 | 5 |
| $MoS_2$ | 1.35 | | 6 | $Ga_2SeTe$ | | 0.21 | 5 |
| $GaPO_4$ | 8.5 | | 7 | $Ga_2STe$ | | 0.25 | 5 |
| α-$In_2Se_3$ | 0.34 | | 8 | $Ga_2SSe$ | | 0.07 | 5 |
| CdS | 32.8 | | 9 | $In_2SeTe$ | | 0.13 | 5 |
| ZnO | 23.7 | | 10 | $In_2STe$ | | 0.25 | 5 |
| ZnO | 21.5 | | 11 | $In_2SSe$ | | 0.18 | 5 |
| Graphene on $SiO_2$ | 14 | | 12 | $MoS_2$ | 1.03 | | 13 |
| Graphene doped with K | | 0.23 | 14 | MoSSe | 0.1 | | 15 |
| Graphene doped with Li | | 0.15 | 14 | $BaTiO_3$ | 8 | | 16 |
| Graphene doped with H | | 0.11 | 14 | PVDF | -36 | | 17 |
| Graphene doped with F | | 0.0018 | 14 | PZT | 7-15 | | 18 |
| Graphene doped with F, Li | | 0.30 | 14 | PbO | 29.6 | 3.9 | This work |
| Graphene doped with H, F | | 0.034 | 14 | | | | |

## References


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