# **Supporting Information**

## Electro-elastic properties of Piezoelectric Te<sub>2</sub>O(PO<sub>4</sub>)<sub>2</sub> Crystal

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#### 1. Calculation formula and process of electro-elastic constants.

The dielectric constants can be obtained by the measured capacitance values according to the following formulas with square plate samples k, l, and j, and circumgyrate sample g:

$$\varepsilon_{ij} = \frac{\varepsilon_{ij}^{T}}{\varepsilon_{0}} = \frac{C_{ij} \times t}{A \times \varepsilon_{0}} (i = 1, 2, 3) \# (1)$$
  
$$\varepsilon_{33}^{T}(\theta) = \varepsilon_{11}^{T} \sin^{2}\theta + 2\varepsilon_{13}^{T} \cos\theta \sin\theta + \varepsilon_{33}^{T} \cos^{2}\theta \# (2)$$

where C is the capacitance, t is the thickness, A is the area of the measured sample, and  $\varepsilon_0$  is the vacuum dielectric constant.

$$s^{E} = \frac{1}{4\rho l f_{a}^{2}(1-k^{2})} \# (3)$$

$$k^{2} = \frac{\pi f_{r}}{2f_{a}} \cot \frac{\pi}{2} \left(\frac{f_{r}}{f_{a}}\right) \# (4)$$

$$d = k\sqrt{\epsilon s} \# (5)$$

$$s^{E} = \frac{1}{4\rho w^{2} f_{a}^{2}(1-k^{2})} \# (6)$$

$$s^{E} = \frac{1}{4\rho t^{2} f_{a}^{2}(1-k^{2})} \# (7)$$

$$s^{E} = \frac{1}{4\rho l^{2} f_{r}^{2}} \# (8)$$

$$\frac{k^{2}}{1-k^{2}} = \frac{\pi f_{a}}{2f_{r}} \cot \frac{\pi}{2} \left(\frac{f_{a}}{f_{r}}\right) \# (9)$$

$$s_{33}^{'}(XZw)\theta = s_{11}sin^{4}\theta + (2s_{13} + s_{55})sin^{2}\theta cos^{2}\theta$$

$$+ 2s_{15}sin^{3}\theta cos\theta + s_{33}cos^{4}\theta + 2s_{33}sin\theta cos^{3}\theta \# (10)$$

$$s_{22}^{'}(XYt)45^{\circ} = (s_{22} + 2s_{23} + s_{44} + s_{33})/4 \# (11)$$

$$s_{11}^{'}(XYl)45^{\circ} = (s_{11} + 2s_{12} + s_{66} + s_{22})/4 \# (12)$$

$$s_{33}'(YZw)45^{\circ}/-45^{\circ} = \binom{4s_{22}+s_{11}+s_{33}+2s_{15}+2s_{13}+s_{55}+2s_{35}}{+4s_{23}+4s_{12}+4s_{25}+2s_{44}+2s_{66}+4s_{46}} / 16\#(13)$$

$$s_{44}(XYl)45^\circ = 0.5s_{44} + s_{46} + 0.5s_{66}\#(14)$$

$$\begin{aligned} d_{13}'(XYw)\theta &= d_{11}\sin^2\theta\cos\theta - d_{31}\sin^3\theta + d_{13}\cos^3\theta \\ &+ d_{33}\sin\theta\cos^2\theta + d_{15}\sin\theta\cos^2\theta - d_{35}\sin^2\theta\cos\theta \# (15) \end{aligned}$$

 $c=s^{-1}\#(16)$ 

Elastic compliance constants  $s_{11}$  and  $s_{33}$  and piezoelectric constants  $d_{11}$  and  $d_{33}$  were calculated using equations (3)-(5) based on samples a and b. When the electric field was applied along the thickness direction of samples c-f, piezoelectric constants  $d_{13}$ ,  $d_{12}$ ,  $d_{32}$ , and  $d_{31}$  and elastic compliance constants  $s_{33}$ ,  $s_{22}$ , and  $s_{11}$  were obtained by equation (5), (8), and (9). Samples c-f were also used to calculate the piezoelectric constants  $d_{35}$ ,  $d_{15}$ ,  $d_{24}$ , and  $d_{26}$  and the elastic constants  $s_{55}$ ,  $s_{44}$ , and  $s_{66}$  by equations (4)-(7), where the electric field was applied along the length direction of the samples. Using the sample g-i and combining equations (8) and (9), the elastic compliance constants  $s_{13}$ ,  $s_{15}$ , and  $s_{35}$  are obtained. Similarly, elastic compliance constants  $s_{23}$ ,  $s_{12}$ , and  $s_{25}$  were obtained in the same way based on samples m-o and equations (8) and (11)-(13). Elastic compliance constant  $s_{46}$  was calculated based on the equations (4), (5), (7), and (14), according to the measurement of the thickness-shear vibration mode of sample p with an electric field applied along the length direction. Samples g and i were also used to verify the sign and value of piezoelectric strain constant  $d_{15}$  according to equation (15). Stiffness coefficients  $c_{ij}$  can be obtained using equation (16).

| Sample  | Modes   | Constants   |
|---|---|---|
| X square plate<br>Y square plate<br>Z square plate<br>(XZw)45° bar<br>X rod   | -<br>longitudinal length extensional              | $\varepsilon_{11}^{T}, \varepsilon_{22}^{T}, \varepsilon_{33}^{T}, \varepsilon_{13}^{T}$ $d_{11}$ |
| Z rod   | vibration mode                                    | <i>d</i> <sub>33</sub>  |
| ZX bar<br>XY bar<br>ZY bar<br>XZ bar  | longitudinal length extensional<br>vibration mode | $s_{11}, d_{31}$<br>$s_{22}, d_{12}, d_{32}$<br>$s_{33}, d_{13}$                                  |
| XZ bar  | transverse length extensional                     | $s_{44}, d_{24}$  |
| XZ bar  | vibration mod                                     | $s_{66}, d_{26}$  |
| XZ bar<br>ZX bar  | thickness shear vibration mod                     | $s_{55}, d_{15}, d_{35}$  |
| (XZw)45° bar<br>(XZw)30° bar<br>(XZw)-30° bar                                 | longitudinal length extensional vibration mode    | \$13, \$15, \$35  |
| (XY <i>t</i> )45° bar<br>(ZX <i>t</i> )45° bar<br>(ZX <i>tw</i> )45°/-45° bar | longitudinal length extensional vibration mode    | <i>S</i> <sub>12</sub> , <i>S</i> <sub>23</sub> , <i>S</i> <sub>25</sub>                          |
| (XY <i>l</i> )45° bar   | thickness shear vibration mod                     | s <sub>46</sub>   |

**Table S1.** The crystal cuts and vibration modes for the determination of electro-elastic constants ofTPO crystal.