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# Homogenization of periodic 1-3 piezocomposite using wave propagation: toward an experimental method

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1-3 piezocomposites are first choice materials for integration in ultrasonic transduc-1 ers due to their high electromechanical performance, particularly in their thickness 2 mode. The determination of a complete set of effective electroelastic parameters 3 through a homogenization scheme is of primary importance for their consideration as 4 homogeneous. This allows for the simplification of transducer design using numeri-5 cal methods. The method proposed is based on acoustic wave propagation through 6 infinite piezocomposite that are considered homogeneous material. Christoffel tensor 7 components for the 2mm symmetry were expressed to deduce slowness curves in sev-8 eral planes. Simultaneously, slowness curves of a numerical phantom were obtained 9 using a Finite Element Method (FEM). Dispersive curves were initially calculated in 10 the corresponding heterogeneous structure. Subsequent identification of the effective 11 parameters was based on a fitting process between the two sets of slowness curves. 12 Then homogenized coefficients were compared with reference results from a numerical 13 method based on fast-Fourier transform (FFT) for heterogeneous periodic piezoelec-14 tric materials in the quasi-static regime. A relative error of less than 2% for a very 15 large majority of effective coefficients was obtained. As the aim of the manuscript 16 is to implement an experimental procedure based on the proposed homogenization 17 scheme in order to determine the effective parameters of the material in operating 18 conditions, it is shown that simplifications to the procedure can be performed and 19 that a careful selection of only seven slowness directions is sufficient to obtain the 20 complete database for a piezocomposite containing square shaped fibers. Finally, 21 further considerations to adapt the present work to a 1-3 piezocomposite with fixed 22

thickness are also presented.

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#### 24 I. INTRODUCTION

Piezoelectric composite materials are used in various applications such as sensors, ac-25 tuators and transducers. Among the many patterns of spatial distribution between the 26 piezoelectric and inert phases, 1-3 connectivity, i.e., aligned piezoelectric rods embedded 27 in a polymer matrix<sup>1</sup> for the corresponding piezocomposite (13PC), can deliver very high 28 performance in the thickness mode<sup>2</sup>, leading to their application in ultrasonic transducers<sup>3</sup>. 29 In order to design transducers integrating 13PCs, numerical methods such as Finite Dif-30 ference Method (FDM) or Finite Element Method (FEM) are commonly used. However, 31 owing to the complexity of the studied system (number of phases, phase arrangement, mi-32 crostructure), tedious meshing is required when performing detailed modeling of 13PC. 33 Moreover, depending on the target application the high precision levels achieved by such 34 calculations are not always necessary. Users of numerical methods preferentially model these 35 transducers with a 13PC simulated as a single-phase homogeneous material with effective 36 properties. This theoretical step corresponding to a homogenization procedure is performed 37 in the framework of the long-wavelength approximation and a subject that was extensively 38 studied during the 1980s and 90s when piezocomposites, in particular, 13PC first became 39 available. Typically, models were initially developed for purely elastic materials and sub-40 sequently extended to take into account the different coupling phenomena, in particular 41 piezoelectricity. 42

<sup>43</sup> The simplest approaches are those of Voigt<sup>4</sup> and Reuss<sup>5</sup> with uniform trial fields. These
<sup>44</sup> have been extended to piezoelectricity to obtain rigorous upper and lower bounds on the

effective free-energy<sup>6</sup>. In 1985, Banno derived an analytical expression for the calcula-45 tion of several effective material parameters for 13PC<sup>7</sup>, based on the model developed by 46 Newnham<sup>1</sup>. Smith  $et al.^2$  in addition, considered thickness mode effective parameters of 47 13PC with analytical expressions. Other workers managed to determine additional effective 48 parameters for all components of the three elastic, dielectric and piezoelectric tensors (elec-49 troelastic moduli) for the different  $piezocomposites^{8-11}$ . However, the accuracy delivered 50 for all parameters related to the thickness mode in these later approaches were not always 51 congruent and very often remained proprietary, in particular parameters for 13PC. 52

At the end of the 1950's, Eshelby had considered the problem of an ellipsoidal inclusion 53 embedded in an infinite matrix and had obtained the analytical expression of the stress and 54 strain fields which turned out to be unchanged within the inclusion<sup>12</sup>. This fundamental 55 result today forms the basis of several mean-field micromechanical models used to estimate 56 the overall elastic moduli tensor. In 1980,  $Deeg^{13}$  extended this solution to the calculation 57 of coupled fields, in particular, piezoelectric materials. Dunn and Taya<sup>14</sup> proposed a method 58 of obtaining effective parameters of a piezocomposite, as did various other workers<sup>15–17</sup>. In 59 these configurations, 13PCs are a particular case of these models where one of the axes of 60 ellipsoidal inhomogeneities tends toward infinity. FEM or more generally numerical methods 61 are also used for 13PC homogenization. The main advantage of numerical methods is that 62 there is no restriction on geometry, size, material parameters and number of phases in the 63 structure<sup>18</sup>. With these methods, different boundary conditions (displacement and electrical 64 potential) are applied to the Representative Volume Element (RVE) in order to determine 65 mechanical and electrical fields inside the material. Details of such methods are given in 66

several reports<sup>19-26</sup>. Recently, the various homogenization methods presented above have
also been adapted and applied to study novel 13PC in order to demonstrate their enhanced
properties<sup>27,28</sup>

For all these models, the required input data are properties of the constituent phases 70 and according to one of the methods briefly described above, this can vary from only a few 71 parameters to the full set of electroelastic moduli for the piezoelectric phase. The effective 72 parameters obtained by these homogenization procedures are therefor directly dependent 73 upon these initial data. However, various 13PC fabrication processes (such as the "Dice 74 and Fill" method<sup>29</sup>) introduce possible variations of properties for each phase and as a 75 consequence variation on the final effective properties. Two examples can be highlighted: 76 the machining process of the piezoelectric phase (starting typically from a bulk ceramic) 77 to design the rods can degrade the piezoelectric properties while addition of the polymer 78 around the rods introduces porosity (air bubbles) which modifies its mechanical properties. 79 Beyond the development of accurate models, measurement of the piezocomposite parameters 80 in operating conditions is essential. Characterization based on 13PC electrical impedance 81 measurements was performed by  $us^{30}$  and the objective of this present work is to develop a 82 homogenization procedure based on acoustic wave propagation. This theoretical procedure 83 was implemented with the understanding that it could then be applied by adapting it to an 84 experimental set-up and thereby effects a correct estimation of homogenized parameters in 85 operating conditions. 86

This approach used mechanical wave propagation through the 13PC and allowed extraction of the effective parameters in a similar way to reported works for different configurations<sup>31–35</sup>. <sup>89</sup> This method as extended to piezoelectric materials, was inspired by Langlet *et al.*<sup>36</sup> who <sup>90</sup> determined the effective tensor of porous elastic materials.

With the assumption of an infinite piezoelectric material, the behavior of wave propaga-91 tion is first presented for the two media of interest in section II. Then, in section III, slowness 92 curves from homogeneous media are fitted on heterogeneous media with the analytical de-93 termination of some elastic parameters before optimization algorithms were used to deliver 94 all the components of the effective electroelastic moduli. From this fit, a homogeneous 95 equivalent medium is determined. In section IV, these results are compared with effective 96 electroelastic coefficients obtained with a Fourier transform-based numerical scheme for pe-97 riodic piezoelectric materials in the quasi-static regime<sup>37–40</sup>. The same initial database for 98 our numerical phantom was used. This comparison was essential and allowed us to validate 99 our new procedure. The influences of two rod shapes and piezoelectric phase volume frac-100 tions on several effective parameters are discussed. Finally, with reference to experimental 101 procedure, two essential points are addressed. First, minimization of the number of slow-102 ness values is examined while simultaneously maintaining accuracy on the deduced effective 103 parameters. Finally, considerations aimed at adapting the present work to a piezoelectric 104 medium with a given thickness and lateral dimensions, is put forward. 105

#### 106 II. WAVE PROPAGATION IN AN INFINITE PIEZOELECTRIC MATERIAL

For a piezoelectric material, constitutive equations interacts the mechanical parameters (strain S and stress T tensors) with the electrical parameters (electric field E and electric displacement D). In the framework of linear behavior, combining Maxwell equations and Hooke's law, the interaction is written as follow<sup>41</sup>:

$$T_{ij} = c^{E}_{ijkl}S_{kl} - e_{ikl}E_{i}$$

$$D_{i} = \epsilon^{S}_{ij}E_{j} + e_{ikl}S_{kl}$$
(1)

where  $c^{E}$ , e and  $\epsilon^{S}$  are, respectively, the elastic tensor at constant electric field, the piezoelectric tensor and the dielectric tensor at constant strain and subscripts  $i, j, k, l \in [1, 2, 3]$ , the 3 space dimensions. Spatial directions 1, 2 and 3 are used indifferently throughout the text with directions X, Y and Z. In the long wavelength approximation, an anisotropic heterogeneous medium can be considered as an anisotropic homogeneous medium if size inhomogeneities are smaller than the selected wavelength<sup>42</sup>. With this assumption, the use of classical plane waves for the study of homogeneous medium in section II B is possible.

#### 118 A. Heterogeneous structure

<sup>119</sup> To study the wave propagation in a 13PC, FEM is used.

#### 120 1. FE model: 4mm- and mm2-structures

In this work, two 13PC designs are investigated. The two were chosen because, from a practical point of view, both can be manufactured by the well-known "Dice and Fill" method<sup>29</sup>. In fig.1, top views of the two periodic configurations are shown with grey piezoelectric square-shaped rods (fig.1.(a))<sup>29</sup> or right-triangle (fig.1.(b))<sup>45</sup> shaped rods surrounded by polymer. To determine the effective symmetry of each structure, the rod-shaped piezoelectric material and the relative positionning of rods to each other, are taken into ac-



FIG. 1. Top view of elementary 13PC patterns for **a**) 4mm and **b**) mm2 symmetries. Surrounded in blue line, are RVEs meshed with FEM with the local orthonormal basis (XYZ). **c**) Unit propagation vector  $\vec{n}$  is defined by angles  $\theta$  and  $\varphi$  in the local basis. Grey areas represent the piezoelectric rods surrounded by a polymer.

<sup>127</sup> count. In the case of Fig1.(a), 4-fold axis of symmetry exists (red square at center of <sup>128</sup> the representation<sup>42</sup>). In the case of Fig1.(b), it is a 2-fold axis (red ellipse at center of the <sup>129</sup> representation<sup>42</sup>). In both cases, two symmetry planes exist (as represented in the structures, <sup>130</sup> fig.1). Accordingly, the square rods structure has 4mm symmetry while the right-triangle <sup>131</sup> rods structure has mm2 symmetry. These symmetries are dependent on the piezoelectric



FIG. 2. Dispersion curves in the mm2-structure for 2 different wave vectors (a)  $\vec{k}(0,90)$ , colinear to  $\vec{k_x}$  and (b)  $\vec{k}(45,90)$ . (c) The final slowness curves for  $||\vec{k}|| = 50m^{-1}$ ,  $0^\circ < \varphi < 90^\circ$  and  $\theta = 90^\circ$ .  $V_{QL}$ ,  $V_{QT1}$  and  $V_{QT2}$  are group velocities for, respectively, quasi-longitudinal, first quasi-transversal and second quasi-transversal modes. Calculation is performed with PZT-4<sup>43</sup> and Epoxy resin<sup>44</sup> for a 1mm-side RVE and a piezoelectric volume fraction  $v_f = 50\%$ .

material used for rods (here, the standard 6mm-symmetry ceramic). Parallelepipedic RVEs of each structure are represented in Fig1 for both cases as blue dotted lines. These RVEs possess the same symmetry as that defined previously for the two structures. To respect the orthotropic axis of 13PC, for the mm2 structure, local basis (OXYZ) is rotated through 45° from the local axis of the 4mm structure. Moreover, the unit propagation vector  $\vec{n}$  is defined by the angles  $\theta$  and  $\varphi$  in the local basis (fig.1.(c)) and  $n_1$  (resp.  $n_2, n_3$ ) is the projection of  $\vec{n}$  on X-axis (resp. Y-axis, Z-axis):

$$n_1 = \sin\theta \cos\varphi, \, n_2 = \sin\theta \sin\varphi, \, n_3 = \cos\theta. \tag{2}$$

For the FEM calculations, ATILA software was used<sup>46</sup>. The whole problem domains 139 are divided into elements connected by nodes, where constitutive equations are locally ap-140 proximated. 1mm-side RVE is chosen. In fact, this is a typical size in XY-plane for 13PC 141 in medical application. The reader is reminded that there is no variation of phases in the 142 z-direction, so the size has no effect on the final results. Isoparametric elements are used 143 with quadratic interpolation along the element's sides. Hexahedrons are used with 20 nodes 144 (8 for the corners and 12 for the middle of the edge) for the 4mm-structure. Similarly, 145 prisms are used with 15 nodes (6 for the corners and 9 for the middle of the edge) for the 146 mm2-structure. Furthermore, the finite element formulation used in ATILA relying upon 147 quadratic interpolation functions, the classical  $\lambda/4$  criterion must be verified in the whole 148 mesh to ensure the validity of the finite element result. The  $\lambda/4$  states that the largest 149 length of each element in a given mesh has to be smaller than a quarter of the wavelength 150 in the material for the working frequency. For structures studied in this paper, the mesh 151 has been chosen in order to respect this criterion. Moreover, the study of periodic 13PC 152 structure is greatly simplified by the Bloch-Floquet theorem because only one RVE needs 153 to be meshed<sup>36</sup>. 154

#### 155 2. From dispersion curves to slowness curves

To establish slowness curves for our two studied structures, properties of PZT-4<sup>43</sup> and Epoxy resin<sup>44</sup> in table II were chosen for the two 13PC phases. Dispersion curves were initially calculated for a 1mm-side RVE with a piezoelectric volume fraction of 50% and only the 3 lowest modes in frequency to be in accordance with the long wavelength approximation. Dispersion curves were calculated for one specific direction of a wave vector in the first Brillouin zone using modal analysis. Wave vector  $\vec{k}$  is defined by  $\vec{k}(\varphi, \theta) = k_x \cdot \vec{n_1} + k_y \cdot \vec{n_2} + k_z \cdot \vec{n_3}$ , where:

163 - 
$$\varphi \in [-180^\circ, 180^\circ],$$

164 
$$\theta \in [0^\circ, 180^\circ]$$
 and if  $\theta = 90^\circ, k \in (XY)$ -plane,

In fig.2, an example of how the slowness curves are determined is put forward for two specific 165 propagation directions in the XY-plane  $(\vec{k}(0,90) \text{ and } \vec{k}(45,90))$  and for the mm2-structure 166 with a step of  $91m^{-1}$  for  $||\vec{k}||$ . Specifically, it is shown in fig.2(a) and fig.2(b), the quasi-167 longitudinal (QL), first quasi-transverse (QT1) and second quasi-transverse (QT2) modes, 168 where QT1 (resp. QT2) mode has an out-plane (resp. in-plane) transverse polarization in the 169 studied plane (here, the XY-plane). A linear behavior exists at low frequency for the three 170 modes where the phase velocity is equal to the group velocity  $(V_{\text{QL}}, V_{\text{QT1}}, V_{\text{QT2}})$ . Calculating 171 dispersion curves for the whole characteristic volumetric region due to mm2 symmetries 172  $(0^{\circ} < \varphi < 90^{\circ} \text{ and } 0^{\circ} < \theta < 90^{\circ})$ , the limit of this linear region is approximatively  $\|\vec{k}\| \simeq$ 173  $1500 \mathrm{m}^{-1}$ . This limit is indicated in fig.2(a) and (b) with the vertical dotted line. To stay 174 within this volumetric linear region,  $\|\vec{k}\|$  was fixed to  $50m^{-1}$  for the rest of the study. In 175 fig. $2(\mathbf{c})$ , final slowness curves (inverse of velocities) are displayed for one quarter of the 176 XY-plane ( $0^{\circ} < \varphi < 90^{\circ}$  and  $\theta = 90^{\circ}$ ). The six velocities calculated from fig.2(a) and (b) 177 are added to slowness curves (crossed points). For FEM calculation, all the slowness points 178 were calculated in this way with a step of  $1^{\circ}$  for  $\varphi$  and  $\theta$ . 179

#### 180 B. Homogeneous structure

From the numerical slowness curves in the heterogeneous material, analytical slowness curves in a homogeneous material had to be established for a further comparison between the two media. A general case is described for the mm2 structure in the following subsection. As in section II A, the medium was considered to be infinite and comprised of piezoelectric material.

#### 186 1. Wave equation in piezoelectric materials

Starting from the expression of the mechanical strain  $T_{ij}$  (eq.1) and removing the electrical potential dependency, the wave equation in a piezoelectric infinite medium can be written<sup>42,47</sup>:

$$\rho \frac{\partial^2 u_l}{\partial t^2} = c_{ijkl}^E \frac{e_{kij} e_{jkl}}{\epsilon_{ik}^S} \frac{\partial^2 u_l}{\partial n_j \partial n_k} \tag{3}$$

where  $u_i$  is the component of the mechanical displacement vector  $\vec{u}$  on the axis  $n_i$  and the time t.  $\rho$  is the density of the medium of propagation. In the assumption of a plane wave propagation problem in an infinite medium in the  $n_j$  direction and for the eq.(3), a solution can be written as:

$$u_{i} = u_{i}^{0} e^{j\omega(t - \frac{n_{j}x_{j}}{V})} = u_{i}^{0} F(t - \frac{n_{j}x_{j}}{V})$$
(4)

where  $u_i^0$  is the wave polarization and V the phase velocity. When this solution is included in eq.3, the wave equation extended to piezoelectric medium becomes:

$$\rho V^2 u_i^0 = (\Gamma_{il} + \frac{\gamma_i \gamma_l}{\epsilon}) u_l^0 \tag{5}$$

where  $\Gamma_{il} = c^{E}_{ijkl}n_{j}n_{k}$ ,  $\gamma_{i} = e_{kij}n_{j}n_{k}$  and  $\epsilon = \epsilon^{S}_{jk}n_{j}n_{k}$ . The  $(\Gamma_{il} + \frac{\gamma_{i}\gamma_{l}}{\epsilon})$  term is called the piezoelectric Christoffel tensor and written as:

$$\overline{\Gamma}_{il} = \begin{vmatrix} \overline{\Gamma}_{11} & \overline{\Gamma}_{12} & \overline{\Gamma}_{13} \\ \overline{\Gamma}_{12} & \overline{\Gamma}_{22} & \overline{\Gamma}_{23} \\ \overline{\Gamma}_{13} & \overline{\Gamma}_{12} & \overline{\Gamma}_{33} \end{vmatrix}$$
(6)

Eq.5 is an eigenvalues equation where the wave polarization  $u_l^0$  is the eigenvector of  $\overline{\Gamma}_{il}$  with its eigenvalue  $\lambda = \rho V^2$ . As  $\overline{\Gamma}_{il}$  is symmetrical, eigenvalues are real and to provide real velocities, eigenvalues must also be positive. Eigenvectors are orthogonal because of the symmetry of  $\overline{\Gamma}_{il}$ . Therefore, solving eq.5 is equivalent to finding the roots of equation:

$$\left|\overline{\Gamma}_{il} - \rho V^2 \delta_{il}\right| = 0 \tag{7}$$

where  $\delta_{il}$  is the Kronecker symbol. Eq.7 has three solutions for a given propagation direction  $\vec{n}$  that are velocities from the 3 known plane waves QL, QT1 and QT2 (i.e., section II A 2).

#### 204 2. mm2-structure (right-triangular rods 13PC)

As mentionned earlier, the most general case is developed in this section. A mm2 piezoelectric material is characterized by 17 independent parameters: 9 elastic coefficients  $(c_{11}^E, c_{12}^E, c_{13}^E, c_{22}^E, c_{23}^E, c_{33}^E, c_{44}^E, c_{55}^E, c_{66}^E, ), 5$  piezoelectric coefficients  $(e_{15}, e_{24}, e_{31}, e_{32}, e_{33}, )$  and 3 dielectric parameters  $(\epsilon_{11}^S, \epsilon_{22}^S, \epsilon_{33}^S, )$ . In comparison, a 4mm-structure is described by 5 elastic, 3 piezoelectric and 3 dielectric independent parameters. For both cases, all components of the piezoelectric Christoffel tensors are analytically determined (Appendix A).

From these expressions, velocities  $V_{QL}$ ,  $V_{QT1}$  and  $V_{QT2}$  can be determined using eq.7 for any

direction. Depending on the symmetry class, calculations for all directions is not always necessary and can be restricted. For a mm2-structure, three planes of propagation are sufficient: XY-plane, XZ-plane and YZ-plane. Solutions for the XY-plane are described in detail here and the two other planes in Appendix B.

In the XY-plane, the components of the projection vector  $\vec{n}$  are given by:

$$n_1 = \cos\varphi , n_2 = \sin\varphi , n_3 = 0 .$$
(8)

A direct consequence of this is the zero value for the  $\overline{\Gamma}_{13}$  and  $\overline{\Gamma}_{23}$  terms in the Christoffel tensor and a simplification of the expression for  $\overline{\Gamma}_{11}$ ,  $\overline{\Gamma}_{12} = \overline{\Gamma}_{21}$ ,  $\overline{\Gamma}_{22}$  and  $\overline{\Gamma}_{33}$  components. Consequently, eq.7 is reduced to:

$$\left(\overline{\Gamma}_{33} - \lambda\right) \left[ \left(\overline{\Gamma}_{11} - \lambda\right) \left(\overline{\Gamma}_{22} - \lambda\right) - \overline{\Gamma}_{12}^2 \right] = 0 \tag{9}$$

where  $\lambda = \rho V_i^2$  are eigenvalues. The three solutions of eq.9 are:

$$\begin{cases} \lambda = \overline{\Gamma}_{33} \\ \lambda_{\pm} = \frac{1}{2} \left[ \overline{\Gamma}_{11} + \overline{\Gamma}_{22} \pm \sqrt{\left(\overline{\Gamma}_{11} + \overline{\Gamma}_{22}\right)^2 - 4\left(\overline{\Gamma}_{11}\overline{\Gamma}_{22} - \overline{\Gamma}_{12}^2\right)} \right] \end{cases}$$
(10)

To assign the solutions to the correct polarizations (i.e., QL, QT1 or QT2 mode), eigenvectors are determined. As an example, one can describe the expression of the simplest solution for the XY-plane is detailed as:  $\lambda = \overline{\Gamma}_{33}$ . Substituting  $\overline{\Gamma}_{33}$  by its definition, the complete expression of the corresponding velocity becomes:

$$V_{QT2}^{2} = \frac{1}{\rho} \left[ c_{55}^{E} \cos^{2}\varphi + c_{44}^{E} \sin^{2}\varphi + \frac{(e_{15}\cos^{2}\varphi + e_{24}\sin^{2}\varphi)^{2}}{\epsilon_{11}^{S}\cos^{2}\varphi + \epsilon_{22}^{S}\sin^{2}\varphi} \right]$$
(11)

Doing the same for  $V_{QT1}$  and  $V_{QL}$  (with the two other eigenvalue expressions), the slowness curves can be analytically calculated for a homogeneous piezoelectric material in the 3 planes of interest for a mm2 symmetry case.

#### 228 III. HOMOGENEOUS EQUIVALENT STRUCTURE

The symmetry class of the equivalent homogeneous structure must be the same as the ini-229 tial heterogeneous structure (here 13PC) and this condition must be taken into account when 230 defining all the components of the  $\overline{c}^E$ ,  $\overline{e}$  and  $\overline{\epsilon}^S$  tensors. Overlined variables are components 231 of the homogeneous equivalent structure. In general, if the symmetry class is unknown, the 232 most general triclinic class is chosen by default. In the present case, a mm2-symmetry class 233 was retained because right-triangular rods are PZT4 with 6mm-symmetry class. The ob-234 jective was to deduce all the effective components of the elastic, piezoelectric and dielectric 235 tensors with a comparison of the slowness curves obtained by the two approaches (hetero-236 geneous and homogeneous materials materials) described earlier. This determination of all 237 the parameters is performed in two steps. The first step involves the direct determination 238 of several elastic parameters. A fitting process of slowness curves involving calculation by 239 FEM is described, in a second step(section IIA). The two step are detailed below. The 240 homogenization process relies on numerical slowness curves calculated by FEM. The case of 242 the mm2-structure (fig.1(b)) with a volume fraction  $(v_f)$  of 50% is presented and numerical 243 slowness curves are presented on fig.3. 244

#### A. Step 1: analytical determination of elastic constants.

For specific directions in any plane (0°, 45°, 90°), the analytical expressions of velocities (section IIB) are simplified to enable the determination of some elastic constants. For instance, in the XY-plane where general equations ( $\varphi$ -dependence) are given by the system



FIG. 3. Numerical slowness curves for three orthogonal planes from the same mm2-structure. Calculation is performed with PZT-4<sup>46</sup> and Epoxy resin<sup>44</sup> for a 1mm-side RVE and a piezoelectric volume fraction  $v_f = 50\%$ .

(10), and when  $\varphi = 0^{\circ}$  and  $\theta = 90^{\circ}$ , velocities are expressed by:

$$\begin{cases} V_{QT2}^{2}(0,90) = \frac{1}{\rho} \left[ \overline{c}_{55}^{E} + \frac{\overline{e}_{15}^{2}}{\overline{\epsilon}_{51}^{S}} \right] \\ V_{QT1}^{2}(0,90) = \frac{\overline{c}_{66}^{E}}{\rho} \\ V_{QL}^{2}(0,90) = \frac{\overline{c}_{11}^{E}}{\rho} \end{cases}$$
(12)

From this set of equations the numerical values from dispersion curves (fig.3(a)) at  $\varphi = 0^{\circ}$ and  $\theta = 90^{\circ}$ ,  $\overline{c}_{11}^E$  and  $\overline{c}_{66}^E$  are easily determined. Similarly,  $\overline{c}_{55}^E$  can be resolved in the case of a purely elastic medium ( $\overline{e}_{15} = 0$ ) but by taking into account the piezoelectricity, three unknowns:  $\overline{c}_{55}^E$ ,  $\overline{e}_{15}$  and  $\overline{\epsilon}_{11}^S$  are added. In a similar way, some elastic constants can be determined for other directions:  $\overline{c}_{22}^E$  in the direction  $\vec{n}(90, 90)$  on the  $1/V_{QL}$  curve and  $\overline{c}_{12}^E$ in the direction  $\vec{n}(45, 90)$  on the  $1/V_{QL}$  and  $1/V_{QT2}$  curves. In general, all elastic constants that can be determined are added to fig.3 in blue color. Other constants that cannot

be analytically and independently determined like  $\overline{e}_{15}$  and  $\overline{\epsilon}_{11}^S$ , are shown in red. Finally, 257  $\overline{c}_{55}^E$  is no longer an unknown value because it can be determined in the  $\vec{n}(0,0)$  direction 258 (XZ-plane) on  $1/V_{QT2}$  curve. The same analysis is performed on the XZ- and YZ- planes 259 (fig.3). Several effective tensor components appear in different directions but in order not 260 to overload fig.3, these are displayed only once. For example, it can be observed that  $\overline{c}_{55}^E$ 261 appears both in the expressions of  $V_{QT2}(0,90)$  (system 12) and  $V_{QT2}(0,0)$  (fig.3(b)). All the 262 six parameters that can be determined analytically are summurized in table I. The same 263 table is applicable for the 4mm-structure, except that the (90,90)-direction and the QT1 264 mode from the (90, 90)-direction are not necessary, for this symmetry structure. 265

Finally, for the mm2-symmetry, with three orthogonal planes and six different directions, a system of nine independent equations is achieved with 11 unknowns. Here, the full determination of tensor components is not analytically possible. Consequently, the second step is a numerical determination performed by a fitting process.

#### B. Step 2: fitting process.

For a full determination of effective tensors, eleven constants have to be fitted:  $\overline{c}_{13}^E$ ,  $\overline{c}_{23}^E$ ,  $\overline{c}_{33}^E$ , all the five constants of  $\overline{e}$  tensor and the three constants of  $\overline{e}^S$  tensor. The fitting process by the objective function (OF) that is minimized and the settings for the optimization algorithm (OA) are described below. Both slowness and velocity are used interchangeably throughout the text because once one is determined, the other one is simultaneously determined too. TABLE I. Analytical expressions for particular directions used for determining six elastic constants in mm2-structure.

	$(\varphi,  heta)$	Mode	$ \rho V_{QL,QT1,QT2} = $				
	(0.00)	$\mathrm{Q}_L$	$\overline{c}^E_{11}$				
e	(0,90)	$\mathbf{Q}_{T1}$	$\overline{c}^E_{66}$				
Y-plan	(90,90)	$\mathrm{Q}_L$	$\overline{c}_{22}^E$				
X			$\frac{A+B}{4} \pm \frac{\left[(A+B)^2 - AB + \left(\overline{c}_{12}^E + \overline{c}_{66}^E\right)^2\right]^{\frac{1}{2}}}{4}$				
	(45,90)	$\mathbf{Q}_L, \mathbf{Q}_{T1}$	with $A = (\overline{c}_{11}^E + \overline{c}_{66}^E), B = (\overline{c}_{22}^E + \overline{c}_{66}^E)$				
-ZX		$\mathbf{Q}_{T1}$	$\overline{c}^E_{55}$				
	(0,0)	$Q_{T2}$	$\overline{c}^E_{44}$				

#### 276 1. Objective function

The aim of this process is to fit the analytical velocities from a homogeneous medium onto numerical velocities considered as the reference from heterogeneous medium. Therefore, one needs to minimize the difference between the numerical velocities  $v_{ij_{\text{FEM}}}$  and analytical velocities  $v_{ij_{\text{Chris}}}$  for all propagation directions of the three planes of interest for the mm2symmetry. Specifically,  $v_{ij_{\text{FEM}}}$  are gleaned from section II A 2 while  $v_{ij_{\text{Chris}}}$  are calculated for each iteration from equations established in section II B 2 with the new set of parameters to be fitted (unknown components of effective tensors). For each new set, a score  $S_a$  is <sup>284</sup> calculated by the OF using the least squares method:

$$S_{a} = \sum_{i=1}^{M} \left( \sum_{j=1}^{N} \frac{(v_{ij_{\text{FEM}}} - v_{ij_{\text{Chris}}})^{2}}{N < v_{ij_{\text{FEM}}}^{2}} \right)$$
(13)

where M is the number of slowness curves selected for the fitting process (M = 5 for the mm2-structure and M = 3 for the 4mm-structure) and N is the number of propagation directions used (with a default of N = 360). Finally,  $\langle \rangle$  is the mean symbol. The five specific curves (fig.4) were specifically chosen because the remaining unknowns that must be determined are involved in their analytical expressions (see red constants on fig3). 4mm-structure requires fewer slowness curves to be fitted because there are fewer effective independent components, in a similar way to the simplification of table I for this case, as explained earlier.

292



FIG. 4. Selected slowness curves (M=5) for the fitting process displayed on one quarter of the

plane ( $\varphi, \theta \in [0^{\circ} 90^{\circ}]$ ).

293

#### 295 2. Optimization algorithm

The OA selects a new set of parameters based on all previous scores returned by OF for the different sets, previously selected. The time needed to reach the best optimization score is called rate of convergence. In order to achieve a fast convergence rate, an OA based on a canonical search is preferred. Here the Nelder-Mead algorithm<sup>48</sup>, also known as the simplex method was used.

In the case of a mm2-structure, the set of effective parameters to be determined is described by the vector  $x = \{\overline{c}_{13}^E, \overline{c}_{23}^E, \overline{c}_{33}^E, \overline{e}_{31}, \overline{e}_{32}, \overline{e}_{33}, \overline{e}_{24}, \overline{e}_{15}, \overline{e}_{11}^S, \overline{e}_{22}^S, \overline{e}_{33}^S\}$ . One of the drawbacks of using this kind of OA is the requirement of an initial vector  $(x_{init})$  to start the process. Values of  $x_{init}$  are set with the matrix method<sup>11</sup> that provides a good approximation for longitudinal tensor (subscripts 3) but a weak approximation of the transverse tensors parameters. It is worth mentioning that these values are calculated as a function of the 13PC volume fraction of piezoelectric phase.

The second setting for the algorithm is the definition of the search-space limits. These are contained in the boundary vectors  $x_{\rm UB}$  for the upper boundaries and  $x_{\rm LB}$  for the lower boundaries. Here, the choice of a large search-space is preferred and boundary vectors are defined by  $x_{\rm UB} = 5x_{\rm init}$  and  $x_{\rm LB} = 0.01x_{\rm init}$ .

In fig.4, the five selected slowness curves for the fitting process are represented only on one quarter of their planes for the case  $v_f = 50\%$ . The curves are normalized to their own maximum to highlight the differences. The black curves that almost coincide with the dotted blue lines are the FEM calculations. The red curves are the initial slowness curves of the OA and were calculated using  $x_{init}$  in table II.

TABLE II. Case  $v_f = 50\%$ . Elastic ( $c^E$  in GPa), piezoelectric (e in C.m<sup>-2</sup>), dielectric ( $e^S$  in  $/\epsilon_0$ ) constants and density ( $\rho$  in kg/m<sup>3</sup>) for PZT4<sup>43</sup> ceramic, Epoxy<sup>44</sup> resin, mm2- and 4mm- structures calculated by the present method and by the FFT method. Initial vector  $x_{init}$  for the OA and relative differences between FFT and present methods are also given. Star (resp. cross and dash) indicates constants analytically determined on first step (resp. non-value parameter and known parameters from others values).

Constants		$c_{11}^{E}$	$c_{12}^{E}$	$c_{13}^{E}$	$c_{22}^{E}$	$c_{23}^{E}$	$c_{33}^{E}$	$c_{44}^{E}$	$c_{55}^{E}$	$c^E_{66}$	$e_{15}$	$e_{24}$	$e_{31}$	$e_{32}$	$e_{33}$	$\epsilon_{11}^S$	$\epsilon^S_{22}$	$\epsilon^S_{33}$	ρ
PZT-4		139	77.8	74.3	-	-	115.4	25.6	-	30.6	12.7	-	-5.2	-	15.1	730	-	635	7500
Epoxy		<b>7</b> 0 4														9			1100
resin		1.84	3.9	-	-	-	-	-	-	-	-	-	-	-	-	3	-	-	1100
Constants		$\overline{c}_{11}^E *$	$\overline{c}_{12}^E *$	$\overline{c}_{13}^E$	$\overline{c}_{22}^E *$	$\overline{c}_{23}^E$	$\overline{c}_{33}^E$	$\overline{c}_{44}^E *$	$\overline{c}_{55}^E *$	$\overline{c}_{66}^E *$	$\overline{e}_{15}$	$\overline{e}_{24}$	$\overline{e}_{31}$	$\overline{e}_{32}$	$\overline{e}_{33}$	$\overline{\epsilon}_{11}^S$	$\overline{\epsilon}_{22}^S$	$\overline{\epsilon}_{33}^S$	$\overline{\rho}$
$x_{ m init}$		×	×	7.56	×	7.56	40	×	×	×	0.0039	0.0039	-0.27	-0.27	9.15	6	6	333	×
mm2	this work	16.04	8.43	8.28	18.85	9.16	41.50	6.90	4.76	4.98	0.0136	0.0195	-0.33	-0.36	8.69	8	12	332	4300
	$\mathbf{FFT}$	16.28	8.58	8.40	19.12	9.37	40.88	7.00	4.85	5.07	0.0112	0.037	-0.33	-0.40	9.07	10	10	326	-
	rel. dif.(%)	1.54	1.81	1.62	1.41	2.35	1.50	1.47	1.85	1.79	19.3	61.9	0.51	11.6	4.25	18.5	12.2	1.71	-
4mm	this work	19.37	6.37	8.64	-	-	40	5.57	-	3.92	0.0075	-	-0.273	-	9.14	5	-	332	4300
	$\mathbf{FFT}$	19.25	6.39	8.66	-	-	40.73	5.53	-	3.9	0.0167	-	-0.353	-	9.08	9	-	332	-
	rel. dif.(%)	0.62	0.31	0.23	-	-	1.81	0.72	-	0.51	76	-	25.6	-	0.66	57	-	0	-

#### 318 IV. RESULTS AND DISCUSSION

Homogenization results are presented for two different cases: (1) results for a specific volume fraction  $(v_f)$  of 50% for 4mm and mm2 13PC are presented in the first section, (2) parameter variation as a function of  $v_f$  are shown in second section. In both sections, all results are compared with the FFT method for validation. The principle of this numerical scheme is briefly recalled in Appendix C.

#### 324 A. Specific case of $v_f = 50\%$

The volume fraction of  $v_f = 50\%$  was chosen because it is a typical 13PC value employed 325 for medical imaging applications. This value allows for the optimization of the thickness 326 coupling coefficient  $(k_t)$  which is an essential parameter for transducer design<sup>3</sup>. Variations 327 of  $v_f$  are possible (typically between 30% and 90%) depending on the desired optimized 328 properties. Fig.4 shows the solution found by the fitting process (blue dotted lines). The 329 maximum difference between the initialization and the FEM is located on the QL wave 330 in the XZ-plane at 62° and its value is 2.98%. After the fitting process, the solution 331 presents a maximum difference on the QT2 wave in the XY-plane, equal to 0.3% (equating 332 to 10 fold reduction when compared to  $x_{\text{init}}$ ). It is also noticeable that the curves from 333 the QT2 wave in the XY-plane was unchanged between  $x_{\text{init}}$  and the solution. In fact, 334 this curve is governed by Eq.11. OA can only modify variables  $\overline{e}_{24}$ ,  $\overline{e}_{15}$   $\overline{\epsilon}_{11}^S$  and  $\overline{\epsilon}_{22}^S$  in this 335 equation as  $\overline{c}_{44}^E$  and  $\overline{c}_{55}^E$  are already determined and considered fixed values. Essentially, the 336 problem is that, in Eq.11, the term  $\frac{\left(e_{15}cos^2\varphi+e_{24}sin^2\varphi\right)^2}{\epsilon_{11}^5cos^2\varphi+\epsilon_{22}^5sin^2\varphi}$  is approximately 500 times smaller 337

than  $c_{55}^E \cos^2 \varphi + c_{44}^E \sin^2 \varphi$ . This means that at least for this case, the variables  $\overline{e}_{24}$ ,  $\overline{e}_{15}$ ,  $\overline{\epsilon}_{11}^S$ and  $\overline{\epsilon}_{22}^S$  are not highly sensitive. As a result, OA had no influence on them.

In table II, all effective tensor components from the solution are given in the case of 340 the 4mm-structure (fig.1(a)) and the mm2-structure (fig.1(b)). For comparative purposes, 341 the same components obtained by the FFT method are also presented. As expected from 342 the previous paragraph, large relative differences appear for  $\overline{e}_{24}$ ,  $\overline{e}_{15}$   $\overline{\epsilon}_{11}^S$  and  $\overline{\epsilon}_{22}^S$  until 62%. 343 However, these differences are not of critical concern because, in practice, these parameters 344 do not affect the expected behavior of 13PC functions either in thickness or flexural modes. 345 To reduce these differences, additional work needs to be performed during OF selection, by 346 for example, adding another slowness curve for the fit or giving extra-weight for this QT2 347 slowness curve. 348

#### 349 B. Results for $v_f$ variations

In this section, the homogenization method is performed on different volume fractions 350 (10%, 30%, 50% and 70%) and compared with the FFT method. In fig.5, the elastic con-351 stants calculated from the present study (black crosses) are well evaluated for the most 352 effective constants (< 2%). In fact, the largest relative difference with the FFT method (red 353 circles) appears to be 6% for  $\overline{c}_{44}^E$  at  $v_f = 70\%$ . This difference is not significant when com-354 pared with the  $\overline{c}_{44}^E$  value of 24% for 4mm-structure (blue crosses). However, these variations 355 of elastic constants show that the shapes have a stronger effect on the constant characteristics 356 of the x-axis  $(\overline{c}_{11}^E)$  as compared with  $\overline{c}_{22}^E$  on the y-axis and  $\overline{c}_{33}^E$  on the z-axis. 357



FIG. 5. Effective elastic constants (in GPa) as a function of piezoelectric volume fraction  $(v_f)$  from the present study represented with the black cross for mm2-structure and blue plus-sign for 4mmstructure. Results from FFT numerical scheme (red circles) are also added for the mm2-structure.

Fig.6 shows homogenized piezoelectric and dielectric effective parameters. The gap between the FFT and the present method on  $\overline{e}_{24}$  and  $\overline{e}_{15}$  was unexpectedly negligible for the reasons already explained in paragraph IV A. However, this difference was pronounced for the higher volume fractions. Details on the constants' sensitivity are extensively covered by Balé<sup>47</sup>.



FIG. 6. Homogenized piezoelectric (in  $C.m^{-2}$ ) and dielectric coefficients (in  $/\epsilon_0$ ) from the present study represented with the black cross for mm2-structure and blue plus sign for 4mm-structure. Results from FFT method is also added with red circle for mm2-structure. Variations are according to volume fraction  $v_f$ .

# C. Simplification for an experimental homogenization: example of the 4mmstructure

In order to apply the present method to an experimental setup, particular effort has to be expended on the acquisition of 13PC slowness curves for the 3 planes of interest. In fact, in the theoretical methodology (general case), 13PC slowness curves were calculated using FEM in all directions at incremental steps of 1°. Unfortunately, measurements made this way, are extremely laborious but also unnecessary. Rather the objective is to find a trade-off between the number of measurements and the accuracy of the deduced effective tensors. The example of a 4mm-structure was chosen here because it is the most frequently
used in practice.

As shown in section III A, specific directions are necessary to determine several constants analytically: for the 4mm-structure in the XY-plane, x-direction and (45, 90)-direction provide the constants  $\overline{c}_{11}^E$ ,  $\overline{c}_{12}^E$ ,  $\overline{c}_{66}^E$  and the z-direction in the XZ-plane (or YZ-plane) brings the  $\overline{c}_{44}^E$  value. Hence, these three directions are retained. Moreover, the fitting process (section III B) for the 4mm-structure has to determine the constants  $\overline{c}_{13}^E$ ,  $\overline{c}_{33}^E$ ,  $\overline{e}_{15}$ ,  $\overline{e}_{31}$ ,  $\overline{e}_{33}$ ,  $\overline{\epsilon}_{11}$  and  $\overline{\epsilon}_{33}$ . These constants appear in two specific directions of the XZ-plane (or YZ-plane):

• z-direction (already retained at step 1) for  $\overline{c}_{33}^E$ ,  $\overline{e}_{33}$  and  $\overline{\epsilon}_{33}$  on the QL mode,

• and (0, 45)- or (90, 45)-direction for the rest on the QL and QT1 modes.

To ensure the validity of the effective parameters obtained using a reduced number of di-383 rections (reduced case), the relative difference between these values and the general case 384 must not exceed 5%. This limit was arbitrarily chosen. Using only the (0, 45)-direction in 385 addition to the three ((0, 90), (45, 90) and (0, 0)) for the analytical determination, results 386 were compared with the general case (45 directions in 2 planes = 90 directions) and, were 387 generally correct for all volume fractions (10%, 30%, 50% and 70%) with relative differences 388 lower than 10% except for  $e_{15}$  due to difficulties explained in section IV A. In table III, the 389 30% volume fraction case is presented because it is the case which exhibits the highest rel-390 ative differences. Only the fitted parameters are presented because parameter determined 391 by analytical method is, by definition, always equal. It is noticeable that the final results 392 deteriorate with relative differences of 93% for the  $e_{31}$  value and 72% for  $e_{15}$ . This also 393

<sup>394</sup> confirms that substantial experimental information is lost, with a reduction in angles. The <sup>395</sup> objective then is to add a few more experimental data to see whether the accuracy of the <sup>396</sup> final results can be improved.

TABLE III. Effective elastic  $\overline{c}^E$  (*GPa*), piezoelectric  $\overline{e}$  (C.m<sup>-2</sup>) and dielectric  $\overline{\epsilon}^S$  (/ $\epsilon_0$ ) constant of 4mm-structure ( $v_f = 30\%$ ).

	general case	reduced cases							
Properties	90 directions	4 direc	ctions	6 directions					
		(rel. di	.ff. %)	(rel. dif	f. %)				
$c_{13}^{E}$	6,01	6,71	(12)	6,00	(0.1)				
$c^E_{33}$	28,08	26,68	(4)	27,87	(1)				
$e_{15}$	3,54e-4	0.0001	(72)	1,78e-4	(30)				
$e_{31}$	-0.14	-0,27	(93)	-0,14	(0)				
$e_{33}$	$5,\!35$	$5,\!52$	(3)	$5,\!25$	(2)				
$\epsilon_{11}^S/\epsilon_0$	5,81	6,08	(5)	$5,\!56$	(4)				
$\epsilon^S_{33}/\epsilon_0$	202	194	(4)	200	(1)				

397 398

For the algorithm, there is a lack of accuracy evaluated on constants calculated from the QL and QT1 modes in (0, 45)-direction. Consequently, two additional directions: (0, 30)and (0, 60)-directions are considered. In this way, the algorithm is better able to reveal the velocity variations in this plane. These results are also shown in table III (6 directions). <sup>403</sup> As expected, relative differences declined with a maximum of 4% (except for the  $e_{15}$  value). <sup>404</sup> With these three cases, the logical trend of increasing accuracy, coupled with the increase <sup>405</sup> in measured directions, are highlighted. Finally, when angle reductions are applied to the <sup>406</sup> experimental setup, the user is obliged to consider choosing a trade-off between the number <sup>407</sup> of measured directions and accuracy of the effective constants.

#### 408 V. CONCLUSION

A method based on wave propagation was successfully implemented to determine the 400 effective electroelastic moduli of 1-3 piezocomposites. Slowness curves in several planes 410 were used to identify all the parameters, whose, Christoffel tensors were analytically ex-411 pressed. This procedure was performed in two main steps. First, several elastic constants 412 (listed in table II with stars) in particular directions were directly determinated using the 413 quasi-longitudinal and two quasi-transversal modes. Second, to complete generation of the 414 effective data (constants without star in table II) a fitting process using the Nelder-Mead 415 algorithm was performed. In this study, two piezoelectric rod shapes (square and right-416 triangle) that can be designed by the well-known "Dice and Fill" method were selected. 417 These two configurations belong to 4mm and mm2 symmetry class, respectively. For the 418 second and most general case, requiring the determination of 17 constants, 10 ( $\overline{c}_{11}^E, \overline{c}_{12}^E, \overline{c}_{13}^E$ , 419  $\overline{c}_{22}^E, \overline{c}_{33}^E, \overline{c}_{44}^E, \overline{c}_{55}^E, \overline{c}_{66}^E, \overline{e}_{31} \text{ and } \overline{\epsilon}_{33}^S$  were obtained with an accuracy of less than 2%. Larger 420 differences were obtained for the two dielectric and two piezoelectric constants ( $\overline{e}_{24}, \overline{e}_{15}, \overline{\epsilon}_{11}^S$ 421 and  $\overline{\epsilon}_{22}^S$  however, these values have limited influence for most applications using thickness 422 or flexural modes. To validate our approach, the results were compared with a Fourier 423

transform-based numerical homogenization scheme for quasi-static conditions. The idea be-424 hind this development was to use this method in *operating conditions* for 1-3 piezocomposite 425 while also taking into account variations in properties, as compared to the original materials 426 database of each phase, appearing in the fabrication process. From a practical point of view, 427 we showed that with only 7 carefully chosen directions of propagation, the entire database 428 of 1-3 piezocomposite with 4mm symmetry can be determined. Future applications can be 429 guided by adaptation of this method to 1-3 piezocomposite for a given thickness and lat-430 eral dimensions. Here, direct measurements of propagative Lamb waves are more suitable. 431 Just as the method described for volume waves, dispersion curves corresponding to the first 432 three symmetric and antisymmetric theoretical Lamb modes will be exploited. Specific elec-433 trode designs on 1-3 piezocomposite will be used to generate modes in a particular direction 434 and scanning laser vibrometer can be used to measure normal displacements at the surface 435 specimen to deduce experimental disperse curve<sup>49</sup>. This new objective is now underway. 436

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#### 439 APPENDIX A:

For a piezoelectric material with mm2-symmetry, piezoelectric Christoffel tensor's components are:

$$\overline{\Gamma}_{11} = c_{11}^E n_1^2 + c_{66}^E n_2^2 + c_{55}^E n_3^2 + \frac{(e_{15} + e_{31})^2 n_1^2 n_3^2}{\epsilon_{11}^S n_1^2 + \epsilon_{22}^S n_2^2 + \epsilon_{33}^S n_3^2},\tag{A1}$$

$$\overline{\Gamma}_{12} = (c_{12}^E + c_{66}^E)n_1n_2 + \frac{(e_{15} + e_{31})n_1n_3(e_{24} + e_{32})n_2n_3}{\epsilon_{11}^S n_1^2 + \epsilon_{22}^S n_2^2 + \epsilon_{33}^S n_3^2},$$
(A2)

$$\overline{\Gamma}_{13} = (c_{13}^E + c_{55}^E)n_1n_3 + \frac{(e_{15} + e_{31})n_1n_3(e_{15}n_1^2 + e_{24}n_2^2 + e_{33}n_3^2)}{\epsilon_{11}^S n_1^2 + \epsilon_{22}^S n_2^2 + \epsilon_{33}^S n_3^2},$$
(A3)

$$\overline{\Gamma}_{22} = c_{66}^E n_1^2 + c_{22}^E n_2^2 + c_{44}^E n_3^2 + \frac{(e_{24} + e_{32})^2 n_2^2 n_3^2}{\epsilon_{11}^S n_1^2 + \epsilon_{22}^S n_2^2 + \epsilon_{33}^S n_3^2},\tag{A4}$$

$$\overline{\Gamma}_{23} = (c_{23}^E + c_{44}^E)n_2n_3 + \frac{(e_{24} + e_{32})n_2n_3(e_{15}n_1^2 + e_{24}n_2^2 + e_{33}n_3^2)}{\epsilon_{11}^S n_1^2 + \epsilon_{22}^S n_2^2 + \epsilon_{33}^S n_3^2},$$
(A5)

$$\overline{\Gamma}_{33} = c_{55}^E n_1^2 + c_{44}^E n_2^2 + c_{33}^E n_3^2 + \frac{(e_{15}n_1^2 + e_{24}n_2^2 + e_{33}n_3^2)^2}{\epsilon_{11}^S n_1^2 + \epsilon_{22}^S n_2^2 + \epsilon_{33}^S n_3^2}.$$
(A6)

### 442 APPENDIX B:

Expression of the six velocities for XZ-plane and YZ-plane according to the solutions of equation 7:

- XZ-plane:

(

$$n_1 = sin\theta$$
,  $n_2 = 0$ ,  $n_3 = cos\theta$ 

445 Solutions are:

$$\begin{cases} V_{QT2}^2 = \frac{\overline{\Gamma}_{22}}{\rho} \\ V_{QL,QT1}^2 \pm \frac{\overline{\Gamma}_{11} + \overline{\Gamma}_{33} \pm \sqrt{(\overline{\Gamma}_{11} + \overline{\Gamma}_{33})^2 - 4(\overline{\Gamma}_{11}\overline{\Gamma}_{33} - \overline{\Gamma}_{13}^2)}}{2\rho} \end{cases}$$
(B1)

- YZ-plane:

$$n_1 = 0$$
,  $n_2 = sin\theta$ ,  $n_3 = cos\theta$ 

446

$$\begin{cases} V_{QT2}^2 = \frac{\overline{\Gamma}_{11}}{\rho} \\ V_{QL,QT1}^2 \pm \frac{\overline{\Gamma}_{22} + \overline{\Gamma}_{33} \pm \sqrt{(\overline{\Gamma}_{22} + \overline{\Gamma}_{33})^2 - 4(\overline{\Gamma}_{22} \overline{\Gamma}_{33} - \overline{\Gamma}_{23}^2)}}{2\rho} \end{cases}$$
(B2)

#### 447 APPENDIX C:

Considering the unit-cell  $\Omega$  of a periodic piezoelectric media, the quasi-static heterogeneous local problem reads,  $\forall \mathbf{x} \in \Omega$ ,

$$\begin{cases} \operatorname{curl}(\operatorname{curl}^{\mathrm{T}} \mathbf{S}(\mathbf{x})) = \mathbf{0}, & \operatorname{div} \mathbf{T}(\mathbf{x}) = \mathbf{0}, \\ \\ \operatorname{curl} \mathbf{E}(\mathbf{x}) = \mathbf{0}, & \operatorname{div} \mathbf{D}(\mathbf{x}) = \mathbf{0}, \end{cases}$$
(C1)

450 with coupled constitutive equations (1)

$$\mathbf{T} = \mathbf{c}^E : \mathbf{S} - \mathbf{e}^T \cdot \mathbf{E}, \quad \mathbf{D} = \boldsymbol{\epsilon}^S \cdot \mathbf{E} + \mathbf{e} : \mathbf{S}$$
 (C2)

<sup>451</sup> and imposed periodicity conditions on the local fields on the boundary of the unit-cell Ω.
<sup>452</sup> Making use of the Green functions method, the solution fields are expressible as coupled
<sup>453</sup> Lippmann-Schwinger equations<sup>39</sup>

$$\begin{cases} \mathbf{S}(\mathbf{x}) = \langle \mathbf{S} \rangle_{\Omega} - \mathbf{\Gamma}^{0} * \boldsymbol{\tau}(\mathbf{x}) \\ \mathbf{E}(\mathbf{x}) = \langle \mathbf{E} \rangle_{\Omega} - \boldsymbol{\Delta}^{0} * \mathbf{P}(\mathbf{x}) \end{cases}$$
(C3)

with  $\Gamma^0$  and  $\Delta^0$  the Green operators corresponding to a uniform reference material with elastic tensor  $\mathbf{c}^0$  and dielectric tensor  $\boldsymbol{\epsilon}^0$ .  $\langle . \rangle_{\Omega}$  indicate the volume average over the unit-cell. 456 The fields  $\boldsymbol{\tau}$  and  $\mathbf{P}$  are given by

$$\begin{cases} \boldsymbol{\tau}(\mathbf{x}) = (\mathbf{c}^{E}(\mathbf{x}) - \mathbf{c}^{0}) : \mathbf{S}(\mathbf{x}) - \mathbf{e}^{T}(\mathbf{x}) . \mathbf{E}(\mathbf{x}) \\ \mathbf{P}(\mathbf{x}) = \mathbf{e}(\mathbf{x}) : \mathbf{S}(\mathbf{x}) + (\epsilon^{S}(\mathbf{x}) - \boldsymbol{\epsilon}^{0}) . \mathbf{E}(\mathbf{x}). \end{cases}$$
(C4)

<sup>457</sup> The solution fields (C3) can be expressed in Fourier space and the problem is solved using <sup>458</sup> an adequate iterative procedure<sup>38,39</sup>. The effective electroelastic coefficients tensors  $\tilde{\mathbf{c}}^E$ ,  $\tilde{\mathbf{e}}$ <sup>459</sup> and  $\tilde{\boldsymbol{\epsilon}}^S$  are defined by

$$\langle \mathbf{T} \rangle_{\Omega} = \widetilde{\mathbf{c}}^{E} : \langle \mathbf{S} \rangle_{\Omega} - \widetilde{\mathbf{e}}^{\mathrm{T}} . \langle \mathbf{E} \rangle_{\Omega} , \quad \langle \mathbf{D} \rangle_{\Omega} = \widetilde{\boldsymbol{\epsilon}}^{S} . \langle \mathbf{E} \rangle_{\Omega} + \widetilde{\mathbf{e}} : \langle \mathbf{S} \rangle_{\Omega}$$
(C5)

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