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Estimation of third order elastic constants of piezoceramics using DC biased impedance measurements

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Abstract: Third order elastic constants of piezoceramics are estimated from electrical impedance measurements by leveraging the dependence of material parameters on a DC bias voltage induced constant mechanical strain. Linear (second order) piezoelectric material parameters are determined for the biased and non-biased sample by solving an inverse problem matching the frequency-dependent electrical impedance of the sample with the output of a simulation model. Estimates for third order elastic constants are derived from these results, enabling simulations with the estimated parameters and thus a comparison with observations.

Keywords: Piezoelectricity, third order elastic constants, acoustoelastic effect, inverse problem, material characterisation

Motivation

Piezoelectric ceramics are used in a wide range of application areas due to their advantageous electromechanical coupling properties. With design processes becoming increasingly simulation-driven, quantitative knowledge of the material behaviour is essential. While the material behaviour can be considered linear in sensory, small-signal applications, non-linear effects occur in the field of high-power acoustics. It includes applications such as ultrasonic cleaning, bonding, and welding, where the systems' behaviour can be considered primarily linear with superimposed nonlinear properties. With these effects typically not considered during the design phase, their influence can result in unforeseen behaviour in the physical system. In an approach to estimate of third order elastic constants of piezoelectric ceramics, this contribution demonstrates how the parameters of a quadratic mechanical material model, as well as the linear dielectric and piezoelectric coupling behaviour, can be identified.

Material modelling

Using tensor notation, the linear elastic behaviour of a rigid medium is modelled as

$$T_{ij} = c_{ijkl} S_{kl}, \tag{1}$$

using Einstein notation [1] where T_{ij} and S_{kl} are the mechanical stress and strain tensors and c_{ijkl} is the elastic stiffness tensor. This description is considered a linearisation of the material behaviour in a specific operating point. It can be expanded in polynomial terms to a quadratic description:

$$T_{ij} = c_{ijkl} S_{kl} + \tfrac{1}{2} c_{ijklmn} S_{kl} S_{mn}, \tag{2}$$

where c_{ijklmn} is the sixth rank tensor describing the quadratic elastic material behaviour. When describing linear material behaviour, Voigt notation [2] is used to reduce the rank of the elasticity tensor from four to two, i.e. from $3 \times 3 \times 3 \times 3$ to 6×6 by exploiting the symmetry of the stress and strain tensors, which are consequently restructured from 3×3 to 6×1. For the quadratic elastic material description, the dimensionality is reduced to [3]

$$T_i = c_{ij} S_j + \tfrac{1}{2} c_{ijk} S_j S_k, \tag{3}$$

with the third rank tensor c_{ijk} with dimensions $6 \times 6 \times 6$ describing the quadratic elastic material behaviour.

Because high-power acoustic devices are typically driven at resonance with comparatively small electric fields, linear relations are used for dielectric ε_{ij} and coupling e_{ij} behaviour, resulting the material equations [4]

$$T_i = c_{ij} S_j + \tfrac{1}{2} c_{ijk} S_j S_k - e_{ji} E_j \quad \text{and} \tag{4}$$

$$D_i = \varepsilon_{ij} E_j + e_{ij} S_j, \tag{5}$$

with the electric field E_i and the dielectric displacement D_i, each with dimensions 3×1.

Polarised piezoelectric ceramics can be modelled as transversely isotropic, coinciding with the 6mm crystal class, which has ten degrees of freedom for a quantitative, linear, undamped material description [5] and can be parametrised as follows:

$$c_{11}, c_{12}, c_{13}, c_{33}, c_{44},$$
$$\varepsilon_{11}, \varepsilon_{33}, e_{15}, e_{31}, \text{and } e_{33}. \tag{6}$$

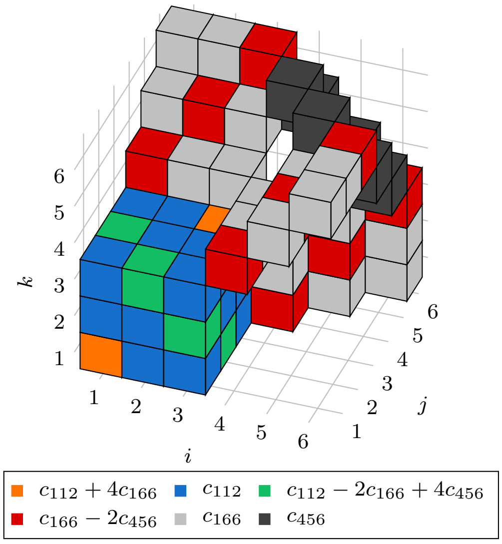

Fig. 1: Visualisation of the non-zero entries of c_{ijk} showing the symmetry of the quadratic elasticity tensor with respect to the indices i, j, and k.

For a transversely isotropic material, the tensor c_{ijk} has nine degrees of freedom [6]. As the purpose of the present study is, however, to model nonlinear effects that are small compared to the linear behaviour, it is assumed sufficient consider isotropic nonlinearity. This reduces the degrees of freedom of c_{ijk} to three, which can be parametrised with

$$c_{112}, c_{166}, \text{and } c_{456} \tag{7}$$

and allows for the application of established relationships from acoustoelastic theory to identify these parameters [7]. Fig. 1 shows a visual representation of the non-zero entries of c_{ijk}. As evident, six different numerical values occur, which can be derived from an arbitrary subset of three values [6, 3].

Experimental procedure

As a basis for the determination of third order elastic constants, linear piezoelectric material parameters are identified from voltage biased electric impedance measurements. This requires an experimental setup that enables the application of a sufficiently high, static voltage while impedance measurements are conducted [8]. Capacitors with adequate dielectric strength and size are used to decouple the bias voltage from the measurement voltage of an impedance analyser. A static voltage of $500\,\mathrm{V}$ is applied, resulting in a static electric field of $500\,\mathrm{V\,mm^{-1}}$ in the sample considered in this study: a hard piezoceramic ring (PIC181 by *PI Ceramic*) with a thickness of $1\,\mathrm{mm}$, and inner and outer radii of $2.6\,\mathrm{mm}$ and $6.35\,\mathrm{mm}$. The electric field is applied to coincide with polarisation direction of the samples. Electric impedance

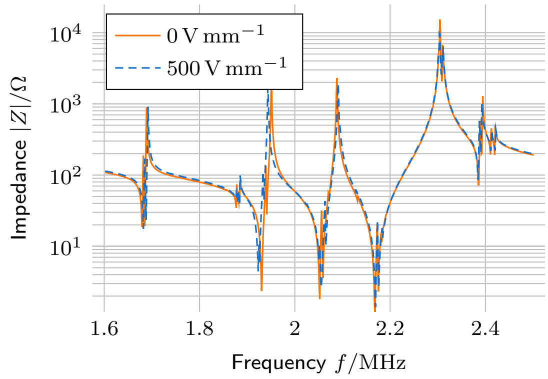

Fig. 2: Section of the electric impedance spectrum of a piezoceramic ring with and without superimposed static electric field.

spectra with frequencies up to $8\,\mathrm{MHz}$ with and without static electric field are acquired. A section with particular sensitivity is shown in Fig. 2. As evident, the influence of the static field can be considered small, however specific resonance frequencies are affected. These impedance spectra are the basis of the subsequent linear material characterisation procedure and consequently for the estimation of third order elastic constants.

Material parameter identification

Based on the impedance measurements, the linear piezoelectric material parameters are identified for the biased and unbiased state by solving an inverse problem. The method applied here is identical to previous work [8]: An axisymmetric finite element model of the piezoelectric sample is implemented in openCFS [9] to acquire synthetic electric impedance spectra for given material parameters. The material parameters are adapted until measurement and simulation are in agreement using gradient-based local optimisation. The initial values required for the optimisation procedure are estimated using a modified convolutional neural network [10]. The results of this identification procedure for the sample in both electric field conditions are listed in Tab. 1 including the parameters of a Rayleigh damping model α_{M} and α_{K}. The density of the sample ($\rho = 7850\,\mathrm{kg\,m^{-3}}$) is measured directly and assumed constant. All parameters show an influence of the static electric field, however, the largest changes are observed in the damping parameters. For the subsequent estimation of c_{ijk} based on the acoustoelastic effect, only the changes in the linear elastic parameters c_{ij} are considered.

Approaches to determine third order elastic constants c_{ijk} based on the acoustoelastic effect are formulated relative to a static stress T_i [7]. In the

Tab. 1: Identified linear parameters for the piezoelectric sample (PIC181) for the biased and unbiased state ($\rho = 7850\,\mathrm{kg\,m^{-3}}$).

Parameter	$0\,\mathrm{V\,mm^{-1}}$	$500\,\mathrm{V\,mm^{-1}}$
c_{11}/GPa	142.17	143.16
c_{12}/GPa	77.42	77.66
c_{13}/GPa	79.49	79.65
c_{33}/GPa	131.04	130.48
c_{44}/GPa	28.29	27.61
$\varepsilon_{11}/\mathrm{nF\,m^{-1}}$	9.39	8.41
$\varepsilon_{33}/\mathrm{nF\,m^{-1}}$	5.90	5.65
$e_{15}/\mathrm{C\,m^{-2}}$	12.33	12.19
$e_{31}/\mathrm{C\,m^{-2}}$	-5.25	-5.33
$e_{33}/\mathrm{C\,m^{-2}}$	14.25	14.03
$\alpha_{\mathrm{M}}/\mathrm{ms^{-1}}$	6.89	11.01
$\alpha_{\mathrm{K}}/\mathrm{ps}$	≈ 0	14.6

present study, no mechanical stress is induced. Instead, the sample shows strain resulting form the applied static voltage via the piezoelectric effect. To still evaluate the measurements using the acoustoelastic relation, the equivalent mechanical stress $T_{\mathrm{eq},i}$, which would result in the same strain is considered. For a given static, electric field E_j ($E_1 = E_2 = 0$ and $E_3 = 500\,\mathrm{V\,mm^{-1}}$), $T_{\mathrm{eq},i}$ is determined using previously identified piezoelectric coupling quantities:

$$T_{\mathrm{eq},i} = e_{ji} E_j, \tag{8}$$

thus assuming homogenous static fields. By exploiting the acoustoelastic relations, which in turn are based on a quadratic material model, the previously mentioned entries of c_{ijk} can be determined [7], where $c_{ij,0}$ and c_{ij} denote the properties of the unstressed and stressed material respectively:

$$c_{166} = \frac{\frac{3c_{11,0}-4c_{44,0}}{T_{\mathrm{eq},3}}(c_{33}-c_{11})+5c_{11,0}+4c_{44,0}-6\frac{c_{11,0}^2}{c_{44,0}}}{6\frac{c_{11,0}}{c_{44,0}}-8}, \tag{9}$$

$$c_{456} = \frac{c_{44,0}}{c_{11,0}-2c_{44,0}}\Big(\frac{3c_{11,0}-4c_{44,0}}{T_{\mathrm{eq},3}}(c_{31}-c_{44,0}) - 4c_{11,0} + 4c_{44,0} - c_{166}\Big), \tag{10}$$

and

$$c_{112} = \frac{1}{2}\Big((c_{33}+c_{11}-2c_{11,0})\frac{3c_{11,0}-4c_{44,0}}{T_{\mathrm{eq},3}} - 3c_{11,0} + 4c_{44,0} - 2\frac{c_{11,0}^2+c_{11,0}c_{116}}{c_{44,0}}\Big). \tag{11}$$

Applying these expressions with the equivalent stress $T_{\mathrm{eq},3}$ yields estimates for the third order elastic constants of the piezoelectric material listed in Tab. 2. The remaining entries of c_{ijk} are determined using the relations visualised in Fig. 1.

Tab. 2: Identified third order elastic constants for the piezoelectric sample (PIC181).

Parameter	Estimate	Parameter	Estimate
c_{166}/GPa	$-2.57 \cdot 10^4$	c_{111}/GPa	$-2.13 \cdot 10^5$
c_{456}/GPa	$7.67 \cdot 10^5$	c_{123}/GPa	$3.01 \cdot 10^6$
c_{112}/GPa	$-1.10 \cdot 10^5$	c_{144}/GPa	$-1.56 \cdot 10^6$

Validation

To assess whether the identified quantitative material model can serve as an adequate approximation of the physical material's dynamic nonlinear behaviour, the following experiment is conducted: The sample is excited with a continuous sinusoidal signal at the first local minimum in the impedance spectrum at $122.6\,\mathrm{kHz}$, which is assumed to be a close approximation for the first radial resonance frequency. The signal is applied with an amplitude of $10\,\mathrm{V}$ and the surface displacement at a point on the centre line of the ring's surface is measured with a laser Doppler vibrometer (VibroFlex QTec by *Polytec*). A displacement signal is recorded after the sample reaches a steady state. The spectrum of the measured displacement (Fig. 3) shows harmonic frequencies occurring at integer multiples of the excitation frequency, indicating nonlinear behaviour.

The described experimental setup is recreated in a 2D nonlinear, time-domain, piezoelectric finite element model assuming radial symmetry [11]. In each time step, the wave equation for elastic waves in solids, considering the nonlinear material behaviour, is solved using an iterative Newton-Raphson approach. The solution of the problem using linear material behaviour is used as initial values for the first time step. All subsequent time steps use the solution of the previous step. Due to the simplicity of the quadratic nonlinearity the Jacobian is calculated analytically. When using a sufficiently short time step in the simulation, the change in the displacement per iteration is small and thus the solution converges after few (typically two) iterations. The displacement simulation result is evaluated at the approximate position it is measured at in the experiment. The simulation is conducted with a sufficient number of time steps for the system to reach a steady state and the spectrum of the displacement is analysed (Fig. 3). The simulation result shows the formation of harmonics at multiples of the driving signal frequency, however, these are significantly less pronounced compared to the measurement results. While the first harmonic is clearly visible in the simulation, it is two orders of magnitude lower compared to the measurement result and would be below the noise floor of a physical system if accurate.

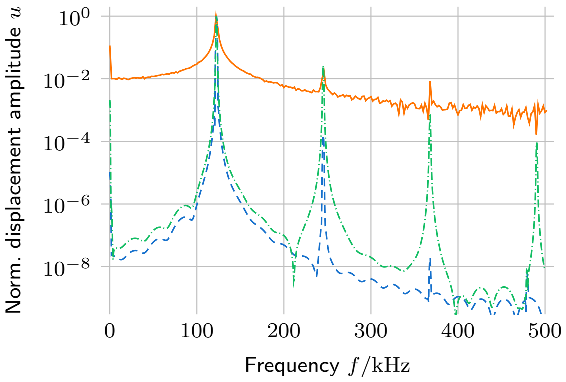

Fig. 3: Spectra of the surface displacement signals resulting from an excitation at the specimens first (linear) radial resonance frequency from measurements (——) and simulations with the estimates for c_{ijk} *(- - - -) and the estimates scaled by a factor of* 100 *(-·-·-).*

The comparison indicates that the proposed approach based on acoustoelasticity clearly underestimates the nonlinear effects occurring in the analysed piezoelectric material. It can, however, be shown that the model is capable of quantitatively reproducing the first harmonic by scaling the nonlinear material parameters up e.g. by a factor of 100 (Fig. 3).

Conclusions

The described method to determine third order elastic constants for piezoceramics based on voltage-biased impedance measurements enables the identification of a quadratic and thus nonlinear elastic material model. However, the estimates provided by the method are shown to be too low to accurately describe nonlinear effects occurring in the sample. One option to rectify these results would be to again solve an inverse problem matching e.g. the simulation results to the measurement (Fig. 3). The deviations are, however, of a magnitude that suggests a revision of the estimation procedure to be more appropriate. An approach would be to assume cubic instead of quadratic material behaviour, which would lead to symmetric properties of the elastic system. A reason for the underestimation of the nonlinearities can also derive from the fact that in the present study, nonlinear effects are exclusive to the elastic behaviour of the material. Taking into account these effects for the dielectric or coupling behaviour can thus be an aspect of future studies.

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References

[1] A. Einstein. "Die Grundlage der allgemeinen Relativitätstheorie". In: *Annalen der Physik* 354.7 (Jan. 1916), pp. 769–822. ISSN: 1521-3889. DOI: 10.1002/andp.19163540702.

[2] W. Voigt. *Lehrbuch der Kristallphysik (mit Ausschluss der Kristalloptik)*. 1966th ed. Vieweg+Teubner Verlag, Jan. 1928. ISBN: 978-3-663-15316-0.

[3] Y. Zhang, J. Jin, and H. Hu. "Third-order elastic, piezoelectric, and dielectric constants". In: *Applied Mathematics and Mechanics* 40.12 (Nov. 2019), pp. 1831–1846. ISSN: 1573-2754. DOI: 10.1007/s10483-019-2550-7.

[4] R. C. Smith. *Smart Material Systems: Model Development*. Society for Industrial and Applied Mathematics, Jan. 2005. ISBN: 978-0-89871-747-1. DOI: 10.1137/1.9780898717471.

[5] W. Heywang, K. Lubitz, and W. Wersing. *Piezoelectricity: Evolution and future of a technology*. Vol. 114. Berlin: Springer, 2008. ISBN: 978-3-540-68680-4.

[6] I. J. Fritz. "Third-order elastic constants for materials with transversely isotropic symmetry". In: *Journal of Applied Physics* 48.2 (Feb. 1977), pp. 812–814. ISSN: 1089-7550. DOI: 10.1063/1.323647.

[7] J. K. Krüger et al. "Nonlinear elastic properties of solid polymers as revealed by Brillouin spectroscopy". In: *Colloid and Polymer Science* 269.8 (Aug. 1991), pp. 764–771. ISSN: 1435-1536. DOI: 10.1007/bf00657442.

[8] O. Friesen et al. "Estimation of piezoelectric material parameters under varying electric field conditions". In: *DAS / DAGA 2025 - 51st Annual Meeting on Acoustics*. 2025, pp. 994–997. DOI: 10.71568/DASDAGA2025.078.

[9] S. Schoder and K. Roppert. *openCFS: Open Source Finite Element Software for Coupled Field Simulation – Part Acoustics*. 2022. DOI: 10.48550/arXiv.2207.04443. arXiv: 2207.04443 [math.NA].

[10] K. Koch et al. "Neuronale Netze zur Startwertschätzung bei der Identifikation piezoelektrischer Materialparameter". In: *tm - Technisches Messen* (2024). DOI: 10.1515/teme-2024-0099.

[11] J. Hölscher. *plutho 0.1.1 – Python Library for coUpled THermo-piezOelectric simulations*. 2025. DOI: 10.5281/zenodo.15608545. URL: https://github.com/emtpb/plutho/releases/0.1.1.