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Impedance measurements for determination of the elastic and piezoelectric coefficients of films

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Abstract

Most of those techniques used for the measurement of elastic coefficients for bulk piezoelectric ceramics are not applicable to films deposited on thick substrates because the measured properties, such as the resonant frequency, are usually dominated by the presence of the thick substrate. This work presents a preliminary study for the application of Alemany et al. automatic iterative method for the determination, from complex impedance measurements, of the film properties using a conventional self-supported cantilever design used in MEMS applications and fabricated from a PZT thick film on a Si-based substrate.

1. Introduction

The resonance method for bulk piezoceramic characterization in the linear range is a commonly used technique developed early in the 20th century, and for which numerous Standards have been issued[1,2]. This method allows the determination of a number of material coefficients, by measuring the frequency dependence of the complex impedance at the electromechanical resonance modes of ferro-piezoelectric ceramics with specific geometries. Iterative[3,4] and fitting methods of the spectra have been developed to provide and alternative to the limitations of the standard calculation methods concerning the characterization of high loss and low sensitivity ferro-piezoelectric ceramic materials.

In the automatic iterative method developed at ICMM-CSIC the material data is determined by solving a set of non-linear equations, having such data as unknowns. To establish these equations, experimental impedance values at a number of frequencies are introduced into the appropriate analytical expression of the wave equation for a given electromechanical resonance mode. This set of equations is established for as many frequencies, which are automatically selected by the program, as unknown coefficients. Solution is carried out by an iterative numerical method[3,4].

For a given resonance mode, such analytical expression is valid for sample geometries with given aspect ratios, which allows the excitation of the uncoupled mode. Regular sample geometries are recommended for the use of this characterization method and extreme care should be taken to determine accurately the dimensions and density of the samples, which are required for the accurate determination of the complex material parameters.

It is worth noting that the measurement of four resonance modes, from three sample shapes (thickness-poled thin disk, thickness-poled shear plate and long rod or bar), are enough to get the full set of independent material parameters (2 dielectric (ε^S_{11} and ε^S_{33}), 3 piezoelectric (d_{33} d_{31} and d_{15})and 5 elastic (s^E_{11}, s^E_{12}, s^E_{33}, s^E_{13}, s^E_{44})), which fully characterizes a ferro-piezoelectric poled ceramic material (6mm symmetry). Iterative methods have been developed for all four resonance modes, which allows a consistent and accurate matrix characterization for this type of material[5].

Since most of the ferro-piezoceramic thin and thick films also have such 6mm symmetry, in principle this method can be extended into film characterization. However, it is not in fact applicable to thin films deposited on thick substrates because the measured properties, such as the resonant frequency, are usually dominated by the presence of the thick substrate and their analytical treatment is not straightforward[6]. This work presents a preliminary study for the application of Alemany et al. automatic iterative method[3,4] to the determination, from complex impedance measurements, of thick and thin film properties, using a conventional self-supported bending cantilever design fabricated with microscale dimensions using MEMS techniques from a PZT film on Si substrate[6].

2. Experimental

2.1.Self-supporting Piezoelectric Cantilevers from Thick Films

The substrate for the fabrication of the freestanding $Pb_{1.2}(Zr_{0.52}Ti_{0.48})O_3$ cantilevers was a 350 μm thick double side polished <100> silicon (Si) wafer, with surface oxide (SiO_2) of 200 nm thickness. The initial processing step was to prepare the wafer for the growth of the piezoelectric layer, this involved the prevention of lead diffusion into the Si wafer through the bottom electrode, which can form a liquid phase lead silicate at the annealing temperatures used for the piezoelectric (~720°C), with the use of a 60 nm thick zirconium oxide layer (ZrO_2). The ZrO_2 was deposited with a sol-gel method where a solution is prepared from 47 ml of ethanol anhydrite at 99.9%, 3ml acetic acid and 4.344 g of zirconium propoxide, all of which are measured under an N_2 ambient atmosphere. The chemicals were then combined and stirred for 60 minutes. The three layers of ZrO_2 each of which were dried at 200ºC and pyrolised at 350ºC, before subsequent crystallisation of the complete ZrO_2 layer was performed in a rapid thermal annealer (RTA) at 800˚C. The wafer was platinised using sputtering in a Nordiko RF/DC sputtering machine, 8 nm of RF sputtered Ti was used as an adhesion layer for the 100 nm thick DC sputtered Pt to act as a the bottom electrode. The thick film lead zirconate titanate (PZT) is composed of a composite slurry containing Ferroperm PZ26 PZT powder mixed with PZT sol to create the bulk of the PZT layer, which due to its high porosity is infiltrated with a PZT sol diluted with 2-Methoxy-Ethanol with a 1:1 ratio to increase densification to 7,07 g/cm^3. The process to deposit a complete layer of PZT uses two depositions of the PZT slurry, with 4 four sol infiltration steps giving a 'layer' thickness of 3.4 µm. Each component of the 'layer' of the PZT slurry and sol infiltration is deposited onto the substrate in a cyclic process which includes drying at 200˚C and pyrolosis 450˚C of the PZT film to provide the required total thickness in our case around 10 µm, prior to sintering in a box furnace at 720˚C. A lift-off lithography process incorporating LOR2A and S1818 resists was used to deposit a patterned top electrode with the same parameters as the bottom electrode. Following the deposition of the top electrode and subsequent lift off, the wafer was RF sputter coated with a blanket layer of gold 200 nm thick, a 2.5 µm thick resist mask was patterned upon the gold layer using the image reversal resist AZ5214E to provide a mould. 1 µm of nickel was then electroplated into the resist mould which was then stripped giving a hard metal mask for the deep reactive ion etching (DRIE) of the PZT film[7] After the etch of the PZT, the hard mask was removed by wet etching the nickel in ferric chloride, and removing the gold in potassium iodide and iodine. The wafer was then patterned using AZ4562 resist to RIE etch the bottom electrode and the surface oxide with Ar and CHF_3, O_2 gasses respectively. After this the wafer was patterned on the back face using the same image reversal technique as the front to allow the sputter deposition and subsequent lift off of a 100 nm thick aluminium layer which acts as the hard mask for DRIE through the Si wafer to release the dies. The final step is the O_2 ashing of the protective resist on the front of the wafer and chemical cleaning using acetone and isopropanol alcohol. The devices obtained for this study are shown in Figure 1.

2.2. Impedance measurements

A custom test rig was fabricated for the purpose of accurate connection to the conducting paths of the devices, enabling testing under reduced pressure below 1bar. Measurements were carried out using an HP4192A LF impedance analyser.

The first fact to take into account for the measurements is that the measuring voltage must be fixed so as to establish a linear response. Figure 2 shows the evolution of the measured resistance and impedance for the L26 device as a function of the voltage applied to the sample. Asymmetric peaks are found for 50mV or higher. Such an effect is well know as to be due to the non-linear behaviour of ferro-piezoceramics under high electric fields and has also been observed in ceramics under 50 V of excitation[8]. Non-linearities are not an issue for ceramic measurements, but must be avoided by use of very low measurement voltage when measuring films.

The second fact that must be considered is the occurrence of coupling between resonance modes. The existence of spurious modes masking the main mode under study must be avoided by carefull choice of the sample dimentional ratio, since the method under study is only valid for single modes. To ilustrate this, Figure 3 shows the a.c.resistance, R, and a.c.conductance, G, obtained from the complex admittance measurements , at the resonances of devices L10 and L26. Whereas the spectrum for L10 shows three peaks, corresponding to three modes of vibration, the L26 device shows an uncoupled unique mode of resonance, amenable for material characterization.

2.3 Identification of the modes of resonance by FEA

The identification of the mode of movement of the resonance used for the parameters calculation is a crucial issue for the validity of the method. To determine the mode of motion, three-dimentional (3D) FEA simulation of the cantilevers was made using the software package ATILA[9], performing 3D harmonic analysis, neglecting the piezoelectric activity. The material parameters were textbook values for the silicon substrate. For the PZT the elastic modulus was calculated based on the measured

density and the average frequency constant of the main resonances for the 3 samples, assuming that these are λ/4 extensional length resonances (Table I).
The FEA model structure of the cantilevers is shown in Figure 4. The length and width of the bar and the width of the substrate where it carries the bar was adjusted to match the different samples. The outer dimensions of substrate and the thicknesses were kept constant.
The results of these simulations of the purely mechanical resonances can be a good guidance for the determination of the modes of motion. However, small deviations in size, mounting of substrate or material parameters can change the behaviour. To validate the results of the FEA simulation, these must be compared with the measured impedance spectrum of the electromechanically driven resonances.
The simulation results for the L10 and L26 cantilevers are shown in Table II.
According to simulation results, both samples have other resonances close to the λ/4 one that is of interest for the calculation of material parameters. However, the vibration pattern for L26 is shaped so the coupling between bar and subtrate is weak, while the coupling is much stronger for L10. Besides, L10 has 3 modes with coupling between the cantilever and the substrate in the frequency range of ±10% around the λ/4 resonance, where L26 has none. FEA simulation also found bending modes of the cantilevers at 2260Hz for the L26, 1205 Hz for L10 and 775 Hz for L28, which are out of the focus of this work.
Figure 3(a) shows the experimental resonance spectra found for the L10 cantilever, with three resonance modes, as shown by the R and G peaks, , in good agreement with the simulated ones. Similarly to the L10 cantilever, the pure length extensional mode of the L28 was found at 292 kHz and a coupled torsional movement with a wavy length extensional mode was found at 325 kHz. Such modes of resonance are very close in frequency and coupling can be expected, thus are not optimum for the calculation of the film parameters by the method studied here. For the L26 cantilever a pure length extensional mode is simulated at 482 kHz. Thus, the measured resonance R and G peaks shown in Figure 3(b) are identified as the length extensional mode of the L26 cantilever.

2.4. Principles for the calculation

Alemany et al. developed an automatic iterative method for piezoceramic characterization that provides the values of ε^T_{33} , s^E_{11} and d_{31}, as well as the electromechanical coupling factor k_{31}, from the impedance measurements at the length extensional mode of a thickness poled long bar. This method is based on the following analytical expression for such resonance mode:

$$Y = G + iB = i\frac{2\pi flw}{t}\left(\varepsilon^T_{33} - \frac{d^2_{31}}{s^E_{11}}\right) + i\frac{2wd^2_{31}}{t.s^E_{11}\sqrt{\rho.s^E_{11}}}\tan\left(\pi.fl\sqrt{\rho s^E_{11}}\right)$$

where f is the frequency of the measurement, t is the thickness of the bar and l,w>>t are the lateral dimensions, and ρ is the density of the piezoelectric material. The details of the method to solve this expression are given elsewhere[3].

This method can also be used for the purpose of characterization from the resonant mode shown in the spectra of Figure 3(b). For the thickness poled cantilever (λ/4 resonator) we have a length extensional mode of motion with a node at one edge, where the cantilever is fixed to the substrate, and maximum amplitude at the other edge. The distribution of strain in the cantilever has been obtained by FEA modelling, the results are shown in Figure 5(a). A freely suspended, thickness poled, bar of double length and, thus, double capacitance of the cantilever one, will have a node at the centre and maximum amplitudes at both ends (λ/2 resonator), as the FEA results in Figure 5(b) shows. Both resonators share boundary conditions and the analytical expression and resonant frequencies are the same for both. Making appropriated geometric corrections it is possible to calculate the material parameters of the cantilever for the mentioned mode using the software of the Alemany method for the length extensional resonance of a thickness poled bar.

3. Results and discussion

The above mentioned calculation was made for the L26 cantilever from the measurements within the linear range using 10 mV input signal, which was determined from measurements using 5 to 100 mV (Figure 2). The results of the calculated parameters and the coupling factor, k_{31} , are shown in Table III. The regression factor, R^2, of the reconstructed spectra using the calculated parameters to the experimental one is high, indicating that the calculus is accurate. However, the high value of ε_{33}^T obtained indicates that admittance data is overestimated. In fact, there is a considerable contribution from a parallel capacitance included in the measurements since the cantilever has an area of 0,33 mm^2 and the conducting path, which is needed to connect the cantilever to the measuring equipment, has an area of 1,5 mm^2 . The size of the parallel stray capacitor (C_{parr}) can be estimated from the measured capacitance outside resonance (C_{tot}) and the area ratio by C_{parr}=C_{tot}.1,5/(1,5+0,33). To correct for C_{parr}, the admittance at a given frequency is then calculated as the measured admittance minus the admittance of C_{parr} at the same frequency.

After this correction was made, we found that the coupling factor increases and there was a lower value of ε_{33}^T at resonance,. The elastic coefficients remained unchanged after the admittance correction.

Conclusions

The feasibility of using the resonance method for the calculation of the dielectric, elastic and piezoelectric complex parameters, as well as coupling coefficients of self-supporting thin or thick films has been shown. The Alemany et al. iterative method for the length extensional resonance mode of thin bars, thickness poled and excited, provide a way to calculate the film complex parameters ε^T_{33} , s^E_{11} and d_{31}, as well as determining the electromechanical coupling factor k_{31}, from an uncoupled resonance of a self-supported thick film cantilevers.

Special care must be taken in the complex admittance measurements concerning the linearity regime of the measurement and the residual capacitance of the connecting pads, which influences both the dielectric and piezoelectric properties determination. Work is in progress to assess the best poling conditions, loading effects upon the top electrodes and the impact of reduced pressure atmosphere for the accurate determination of the material parameters of thin and thick films.

Acknowledgements

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Figure 1. Devices obtained for the study. Film thickness is 10 µm, width 250µm and length 2500, 2000 and 1500 µm (from left to right).

Figure 2. Resistance, R, and conductance, G, measured at the resonance of the L26 cantilever as a function of the measuring voltage.

Figure 3. Resistance and conductance measured at the resonances of the (a) L10 and (b) L26 cantilevers.

Figure 4. Structure for the FEA model of the cantilevers.

Figure 5. Strain distribution at two equivalent resonance modes: (a) the $\lambda/4$ length extensional mode of a thickness poled cantilever of thickness t, width w and length l and (b) the $\lambda/2$ length extensional mode of a thickness poled bar of thickness t, width w and length 2xl.

Table I. Cantilever parameters used for the determination of PZT E-modulus for the FEA modelling.

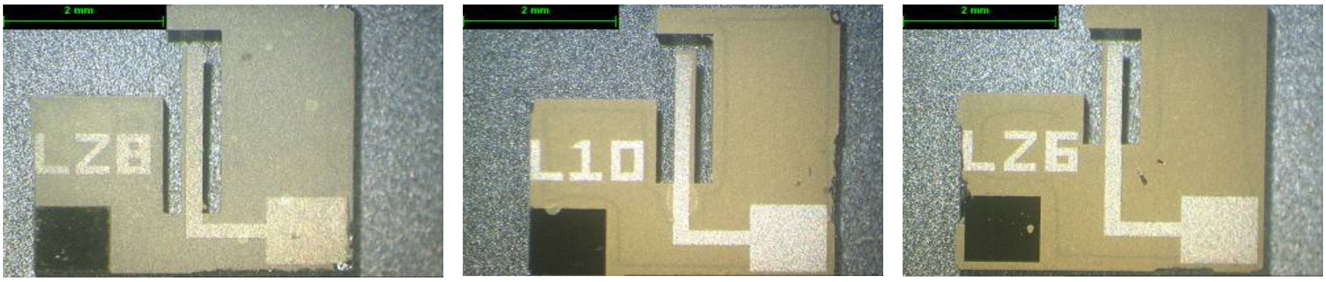
Cantilever	Length (µm)	Measured Resonance Frequency (KHz)	Frequency Constant λ/4 resonator (kHz.mm)	Frequency Constant Deviation (%)
L26	1500	487	731	-0.3
L10	2000	363	725	-1.1
L28	2500	297	743	+1.4
		Average	733	

Table II. Simulated resonance frequencies for the L10 and L26 cantilevers.

L10			L26		
Resonance Frequency (kHz)	Deviation from λ/4 resonance at 366 kHz (%)	Coupling to substrate	Resonance Frequency (kHz)	Deviation from λ/4 resonance at 482 kHz (%)	Coupling to substrate
328	-10	yes	454	-6	no
331	-10	no	468	-3	no
355	-3	yes	475	-1	no
366	**0**	**no**	**482**	**0**	**no**
369	1	no			
373	2	no			
385	5	yes			
403	10	no			

Table III. Resonance characteristics and thick film parameters of L26 device .

PARAMETERS RELATED TO THE FUNDAMENTAL LENGTH EXTENSIONAL RESONANCE MODE OF CANTILEVERS, THICKNESS POLED AND EXCITED	Raw measured data L26 sample measured at 10 mV	Corrected data L26 sample measured at 10 mV
F_s (kHz)	493.82	493.82
F_p (kHz)	494.03	495.08
Auxiliary Frequencies	F1= 486.26 kHz F2= 501.74 kHz	F1= 485.52 kHz F2= 499.07 kHz
Number of iteracions	491	33
R^2	0.999937	0.999915
$\|K_{31}\|$	3.13 %	7.71 %
N_{31} (kHz.mm)	1481	1482
s_{11}^E (10^{-12} m^2N^{-1})*	**16.10 - 0.05 i**	**16.10 - 0.05 i**
d_{31} (10^{-12} $C.N^{-1}$)	**-18,4 + 0.7 i**	**-13.0 + 0.5 i**
ε_{33}^T	**2415 - 58 i**	**200 - 1.8 i**



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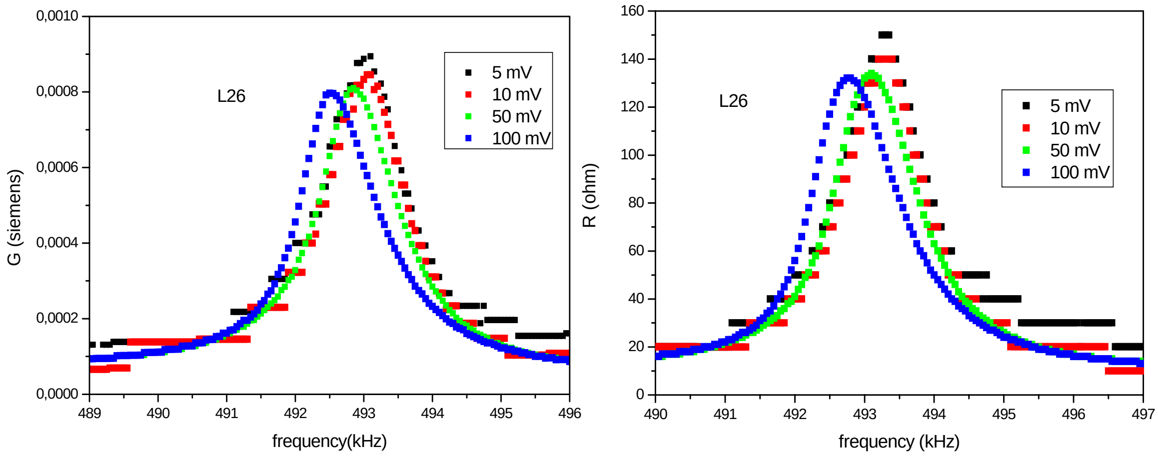

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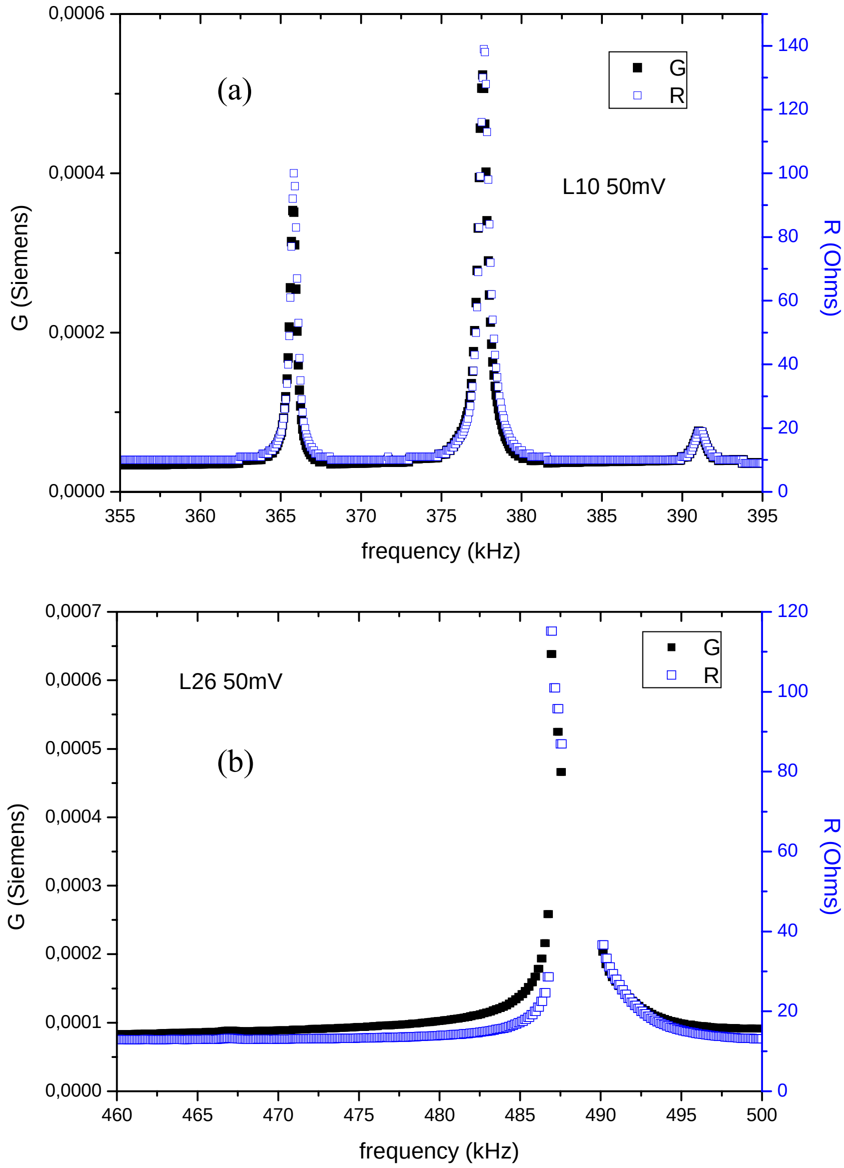
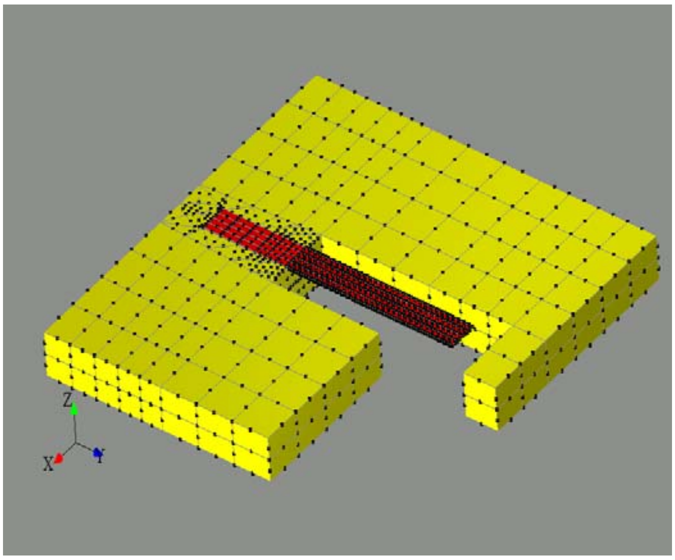


Figure 3. Resistance and Conductance measured at the resonances of the (a) L10 and (b) L26 cantilevers.



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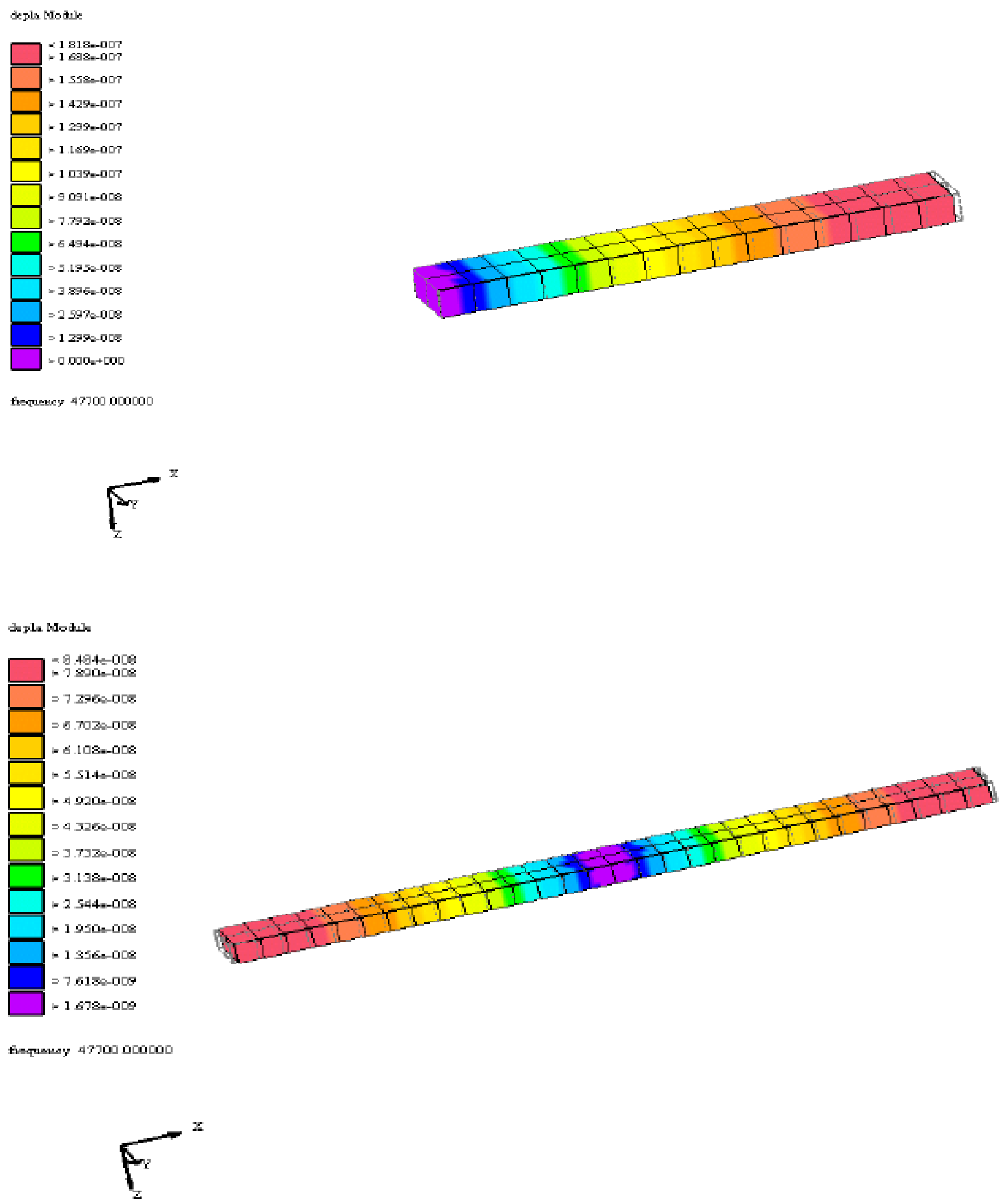

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