

VILNIUS UNIVERSITY  
STATE SCIENTIFIC RESEARCH INSTITUTE CENTER FOR PHYSICAL  
SCIENCES AND TECHNOLOGY

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**DEVELOPMENT OF THE METHODS  
OF BROADBAND DIELECTRIC SPECTROSCOPY  
BY INVESTIGATING  
(1-x)(Na<sub>1/2</sub> Bi<sub>1/2</sub>)TiO<sub>3</sub> - xLa(Mg<sub>1/2</sub> Ti<sub>1/2</sub>)O<sub>3</sub>  
AND OTHER MATERIALS**

Summary of the thesis  
Physical science, physics (02P)

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TECHNOLOGIJOS MOKSL CENTRAS

Saulius Rudys

**PLAIAJUOST S DIELEKTRIN S SPEKTROSKOPIJOS  
METOD TOBULINIMAS, TIRIANT  
(1-x)(Na<sub>1/2</sub> Bi<sub>1/2</sub>)TiO<sub>3</sub> - xLa(Mg<sub>1/2</sub> Ti<sub>1/2</sub>)O<sub>3</sub>  
IR KITAS MEDžiAGAS**

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## Introduction

Dielectric spectroscopy takes important place within physical properties characterisation methods for ferroelectrics and related materials. Even though it is not possible to investigate local structure of material, as using, for example, magnetic resonance methods or atomic force microscopy, one can use dielectric spectroscopy as an excellent indicator of collective response for various materials because of its very wide temperature and frequency range.

Properties of the material can be measured by different methods. Choice of particular method depends on frequency and temperature ranges, value of parameter under observation, dimensions of the sample and available measuring device. A method of measuring is entirety of measuring device, measuring circuit in which the sample is placed, and mathematical model which links readings of the device with properties of the measured material. Obviously, it is not a trivial task to measure dielectric or magnetic properties in a wide frequency range, especially if frequency or dielectric permittivity is high. In this case, requirements for precision in production of sample and measuring circuit are higher than usual. When choosing the method for measuring, one usually aims for highest measurement accuracy, highest frequency and temperature ranges, convenience in preparing the sample and low price. Unfortunately, these requirements are contradictory. Methods for dielectric spectroscopy are progressing together with technologies of measurement devices and computational techniques. Contemporary network analysers cover very wide frequency range - from megahertz to hundred gigahertz. These devices have coaxial connectors, thus it is desirable for measuring methods to be compliant with broadband coaxial lines. Therefore, the open ended coaxial line method is very perspective.

Because of gains in computational power of computers, digital methods of solving Maxwell equations have become applied more widely. They enable us to calculate S parameters of complicated circuits. We are now able to use universal software for electromagnetic modelling which according to user generated three dimensional structure calculates both scattering parameters, and currents and fields distribution in space. In order to calculate properties of the material, which is included into structure under analysis, one needs to solve reversal problem using some kind of iteration

method. This takes a lot of time using even contemporary computers, thus development of analytic (or other) methods requiring less computational power is very relevant task. Universal modelling software is irreplaceable for checking already developed mathematical model or for figuring out the influence of some parasitic circuit parameters to the results of the measurement without creating separate mathematical model.

### **Statements presented for defence**

1. Using multimode capacitor in coaxial line model, it is possible to measure magnetic permeability when dielectric permittivity is high (1-2 order bigger in magnitude than magnetic permeability).
2. The modified open ended coaxial line model is precise and suitable for dielectric permittivity measurements of limited sized samples.
3. The Meyer-Neldel rule for electric conductivity is valid in  $(1-x)(\text{Na}_{1/2} \text{Bi}_{1/2})\text{TiO}_3 - x\text{La}(\text{Mg}_{1/2} \text{Ti}_{1/2})\text{O}_3$  (NBT-LMT) ceramics.

### **The novelty of obtained results**

1. The commercial electromagnetic software was used in unconventional way to calculate materials' electrical properties. Because the software is not adopted for this task, the built-in optimisation option was used. In this way dielectric and magnetic properties of the materials in very complex shape were calculated.
2. Mathematical model of rectangular rod in the waveguide and new model of multimode capacitor were checked by numerical methods.
3. The method to measure small magnetic permeability using capacitor like sample in coaxial line when dielectric permittivity is much higher than magnetic permeability was suggested.
4. The modified open ended coaxial line method for finite dimensions samples was developed and checked by numerical methods.

5. The Meyer-Neldel rule validation in NBT-LMT ceramics group was shown.

### Outline of the thesis

The dissertation consists of the short introduction and 4 chapters, each followed by reference section. Conclusions presented after chapter 3 and chapter 4. The scientific novelty and main statements to be defended are presented in **Introduction**.

**Chapter 1** covers the description of main measurements methods of dielectric spectroscopy in radio frequency range. The open ended coaxial line method is presented in more detail.

**Chapter 2** is devoted to overview the investigation of NBT based solid solutions ceramics. Information about NBT-LMT structure and properties as ferroelectric relaxor is presented.

**Chapter 3** is about investigated measurement methods. The measurements of the permittivity and permeability consist of three main components: measurement equipment, investigated material (in the device under test) and mathematical model. The model relates electrical values measured by the equipment and physical properties of the material, the latter being the goal of the measurement. The mathematical model is simple only in very few cases: a thin capacitor with dielectric between plates, or a transmission line in a dielectric media. However, when frequency is high or permittivity is relatively high, there are difficulties to make device under test in small dimensions and various spurious effects appear, thus simplest models are not acceptable. Other models can be very complex and difficult. Thus, the development of the measurement's mathematical model may be a serious problem. Furthermore, when the geometrical shape of the sample is complicated, no analytical solution can be obtained from Maxwell equations. In such cases, only numerical solutions can be helpful.

Recently, the calculation power of the computers significantly increased. The commercial software, based on various numerical Maxwell equations solution methods appeared which allows EM simulations of complex 3D structures. For example, it is possible to calculate S parameters of the device, when its dimensions and shape and electrical properties of the material are known. When doing dielectric properties

measurements it is necessary to solve the reverse task: to find electrical properties using measured S parameters. However, such software is not adapted for this task.

Some of the EM simulation software has optimization option. It means that if the optimization goal such as S parameter is defined, the program will select device dimensions and (or) electrical properties. In our case the sample, which is a part of the device under test, electrical properties. Software Ansoft HFSS, version 11 was used for our calculations.

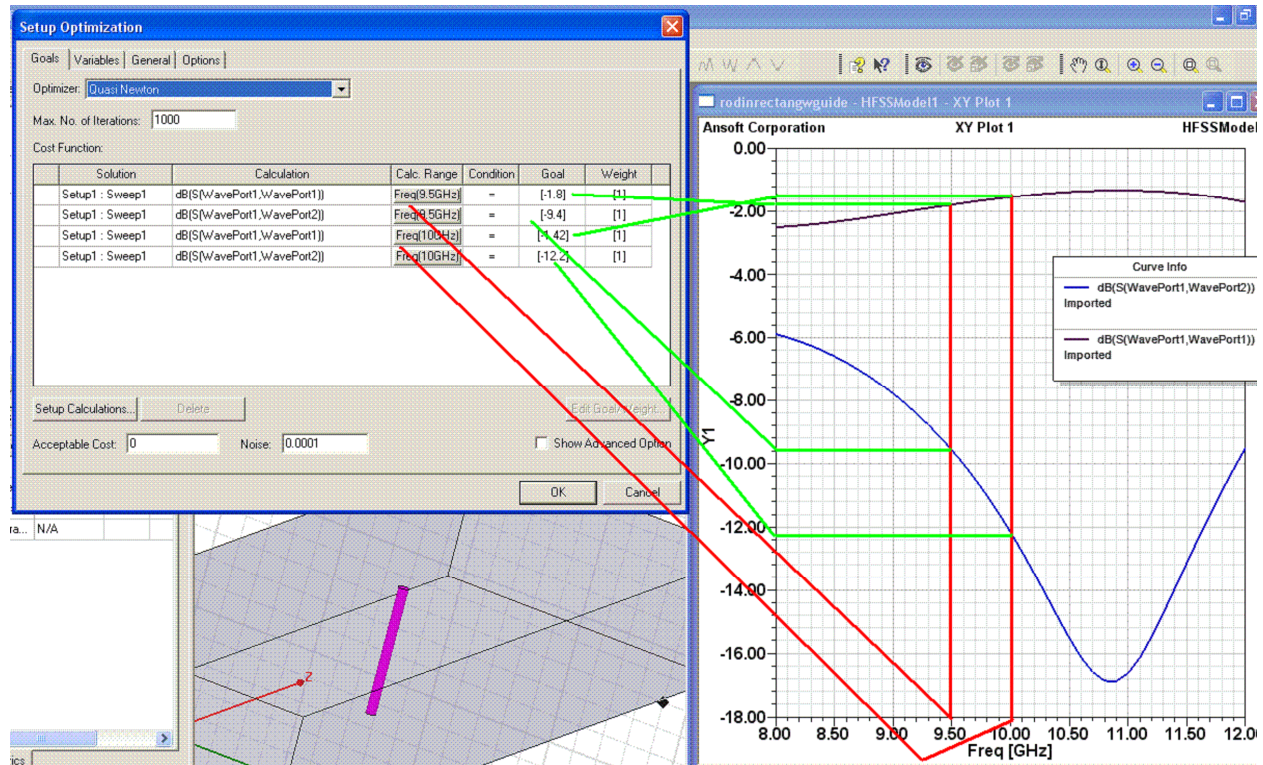


Fig. 1. Optimization setting sample.

In Fig. 1 a sample optimization setting, when device under test is a dielectric rod in a waveguide is shown. Solid curves are experimental reflection and transmission coefficients of the cylindrical rod in the rectangular waveguide. We must mark, that the same values of reflection and transmission may be obtained with different permittivity values. In such case it is much better to define electrical parameters at least at two different frequencies. It allows solving the problem unambiguously.



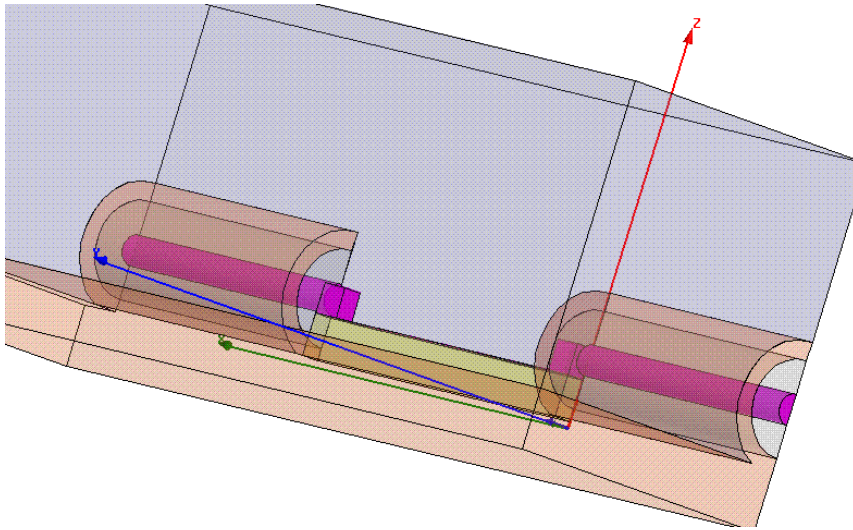


Fig. 2. Microstrip line measurement setup.

Another example is the rectangular dielectric rod with both opposite sides metallization, as shown in Fig. 2. Sample is connected to analyzer by coaxial line, sample is placed on conductive plate, and metallization has finite thickness and conductivity. Values of dielectric permittivity used were 140 for real part and 8 for imaginary. At low frequencies this device can be considered as a microstrip line. At high frequencies inline dispersion, influence of discontinuities of coaxial to microstrip transition appears and microstrip model is not acceptable any more for the calculations. It is very complicated problem to develop and to solve such model by usual analytical methods.

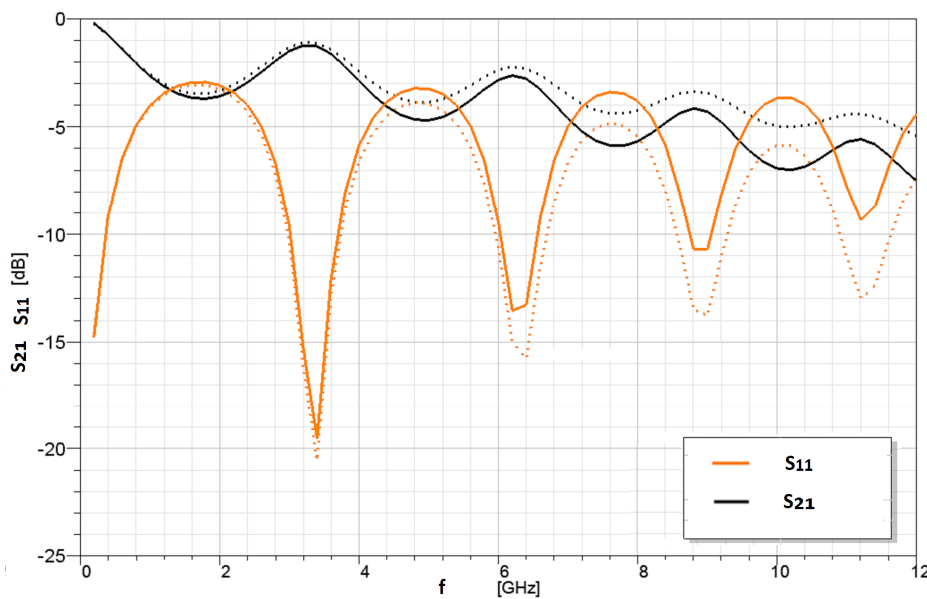
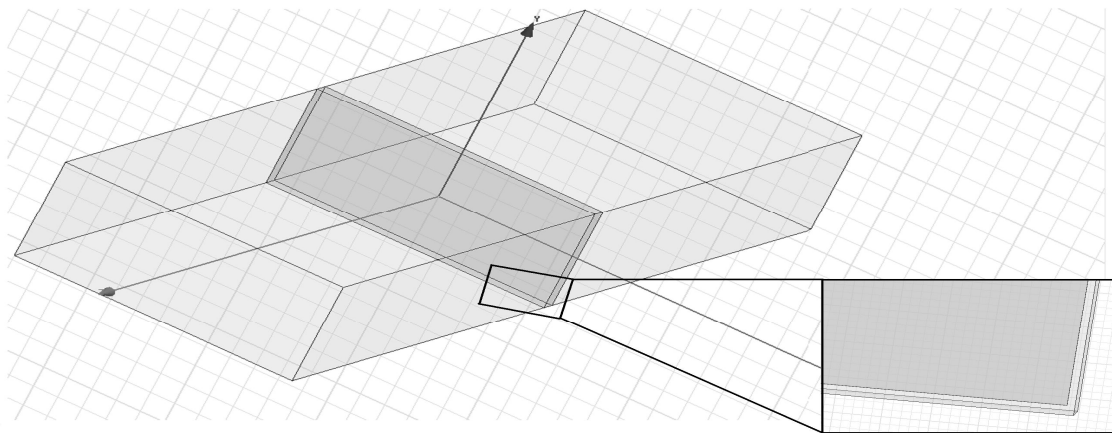


Fig. 3. Calculated  $S$  parameters of the real samples of the geometry presented in the Fig. 2 (solid lines) and ideal samples connected to lumped ports (dotted lines).

Using EM simulation software with optimisation option it is possible to calculate dielectric permittivity from measured S parameters, both at low and high frequencies. By using numerical methods, there is no difference what structure is under simulation ó complex or simple. Difference will be only in computation time. On Fig. 3 calculated S parameters of the real and ideal samples when lumped ports are connected between conductive surfaces at the narrow ends (dotted lines) are shown. Due to various spurious factors described above, there is no difference between realistic and ideal model only at low frequencies. This method has been applied for the microwave measurements of microwave properties of pyrochlore  $\text{Bi}_{1.5}\text{ZnNb}_{1.5}\text{O}_{7-x}\text{F}_{2x}$  (BZN) ceramics [1]. This method was also tested by determining complex dielectric permittivity and complex dielectric permeability of carboncoated capsules of nickel (Ni@C) embedded into polyurethane matrix. At the frequencies 0.05-3 GHz microstrip method like in case of BZN ceramics was used. At frequencies 26 ó 36 GHz classical dielectric filled waveguide method was used [2]. Due to problems in sample fabrication, a gap between sample and waveguide walls appears as in Fig. 4.



*Fig. 4. Dielectric filled waveguide. The gap between wall and sample is shown in the inset.*

Thus the filled waveguide model is not acceptable and it is necessary to take into account this problem doing permittivity and permeability measurements. To solve this problem numerical method was used. As shown in Fig. 5, the gap presence is critical to measurements of the real part of dielectric permittivity.

Summary of the microstrip and waveguide methods used for Ni@C permittivity and permeability measurements is shown in Fig. 6.

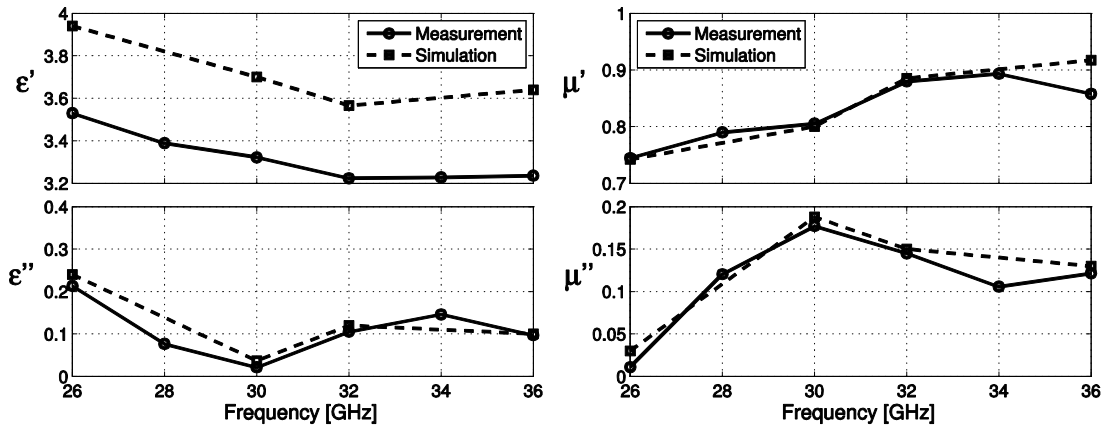


Fig. 5. Dielectric permittivity and magnetic permeability measurements of Ni@C sample with gap using classical model without gap (solid lines) and numerical simulations (dashed lines).

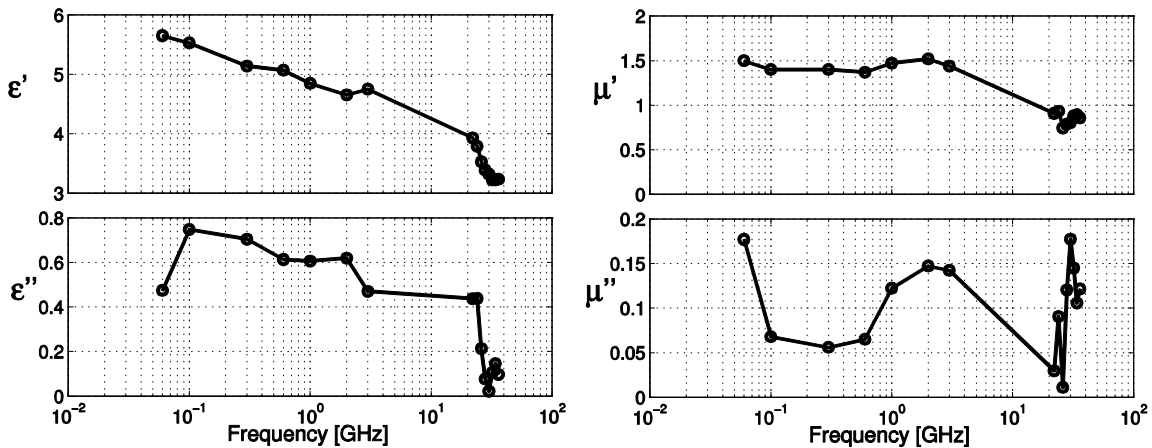


Fig. 6. Summary of the microstrip and waveguide methods used for Ni@C dielectric permittivity and magnetic permeability measurements. At the frequencies 0.05-3GHz microstrip method was used. At frequencies 21 ó 36 GHz waveguide method was used.

Mathematical model on multimode approach and software for dielectric permittivity and S parameters calculation have been created for the case of squared rod in the rectangular waveguide. This model was checked using HFSS software. Excellent agreement between different methods is obtained in Fig. 7.

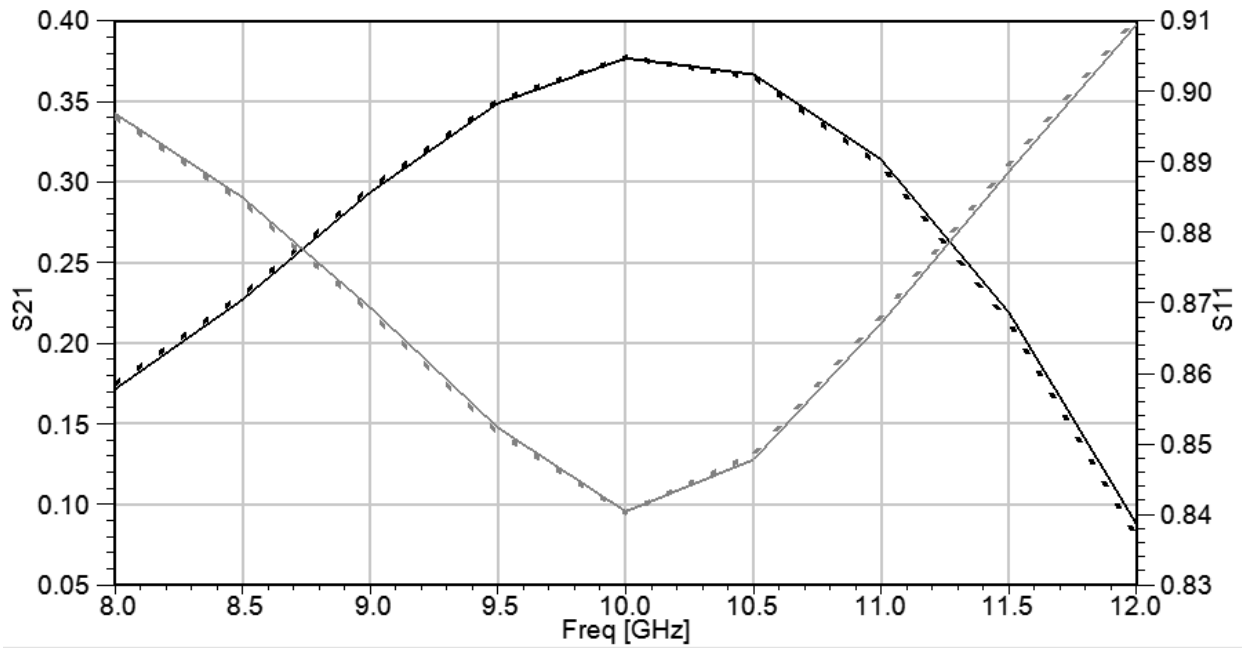


Fig. 7. Calculated by HFSS (solid lines) and multimode model (dotted lines) reflection (black lines) and transmission (grey lines) coefficients.

The measurements of the broadband dielectric spectroscopy of various solid solutions of relaxor ferroelectrics are very complicated due to the high values of dielectric permittivity and dispersion, which extends up to far GHz range. Usually such measurements are performed in coaxial line. At higher frequencies the quasi-static approach fails and one has to use a rigorous approach. The electric field in the coaxial line becomes non-homogeneous, what results in false permittivity calculations. There are some models taking into account field non-homogeneity. According to such models, the wave between capacitor plates is also TEM ( Fig. 8). Fields outside capacitor are not taken in to account. Even using this formalism does not allow to measure dielectric materials, especially ferroelectrics up to GHz frequencies.

The aim of the present work is to improve measurement method of ferroelectrics up to 40 GHz in coaxial line, allowing measurements of ferroelectrics with high dielectric permittivity. It is assumed that dimensions of the coaxial line  $R_1$  and  $R_2$  (Fig. 9) are such that only main (TEM) mode can propagate at the measuring frequencies like in dynamic capacitor model.

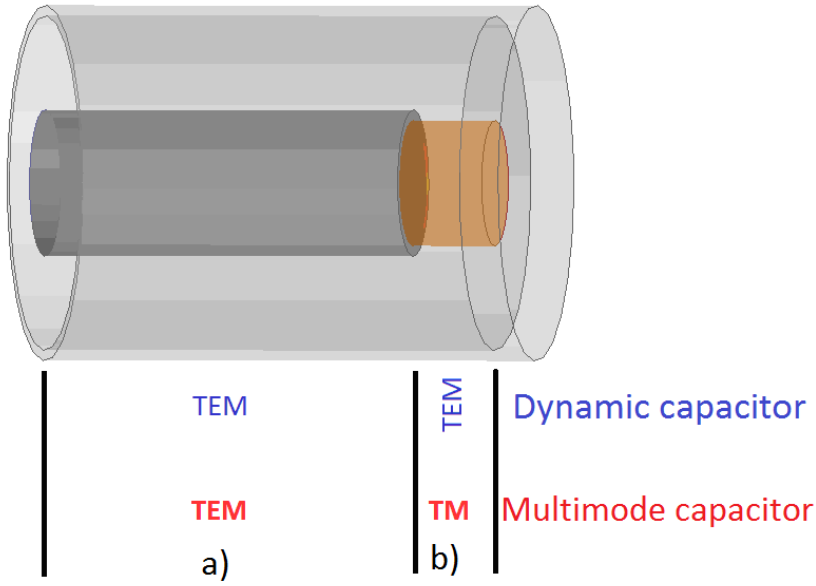


Fig. 8. Difference between dynamic capacitor and multimode capacitor approach in propagating wave type.

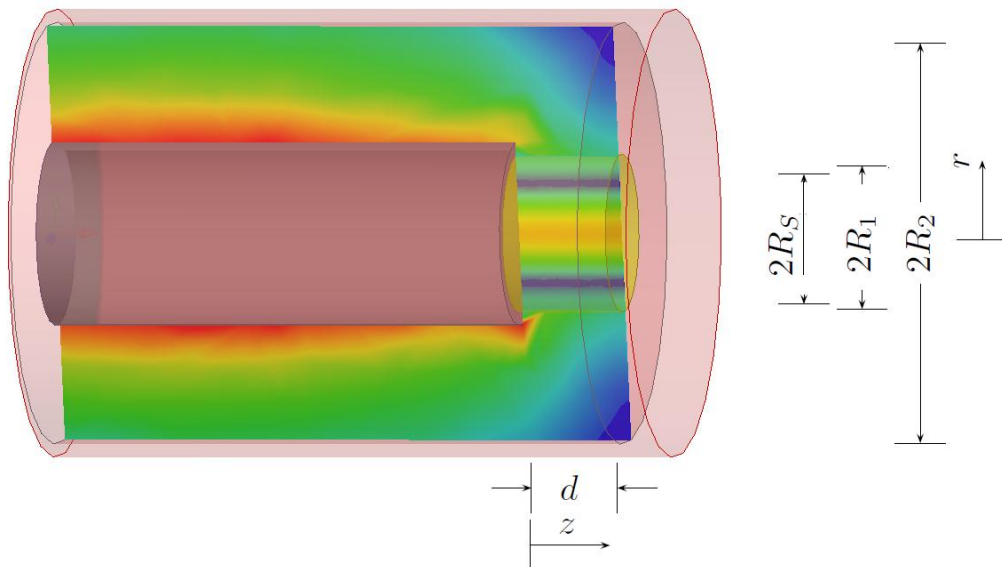


Fig. 9. Structure dimensions of the capacitor model. Electric field distribution (calculated by HFSS) in the capacitor on high frequency (3 GHz).  $\phi=400$ ,  $\delta=40$ ,  $d=4\text{mm}$ ,  $R_s=3\text{mm}$ .

To obtain accurate relations between dielectric permittivity and reflection coefficient we consider the measurement setup as consisting of two regions: a) coaxial line, b) non-homogeneously filled circular waveguide of the length  $d$  consisting of the measured sample, the outer conductor of the coaxial line and terminated by the conducting plane -

short circuit (Fig. 9). The reflection factor is a combination of scattering from the interface between the regions a) and b) and reflection from the terminating plane.

There are several methods to solve this electrodynamics problem. A relatively simple one would be the frequency-domain finite-difference (FD) or finite-element (FEM) method to solve the Maxwell equations on cylindrical coordinate system with the axial symmetric boundary conditions. While easy to implement, the FD method is quite resource (both memory and processor) intensive (depending on the required precision), and the direct problem  $R^*(*)$  takes considerable time to complete. Since we are interested in the reverse problem  $*(R^*)$ , which in non-trivial cases can only be solved by iteration, the efficiency of the method is very important. Therefore we are interested to have a most efficient method albeit more complex analytically.

Here the mode-matching approach comes to mind. It involves dividing an irregular structure into several regular parts (we already have done that), for which the electrodynamics equations can be solved analytically in terms of propagation modes. The mode amplitudes are then obtained by matching electric and magnetic fields on the interfaces between the parts. The general idea is that only a small number of lowest-order modes are often required to obtain sufficient accuracy, thus reducing the amount of calculations.

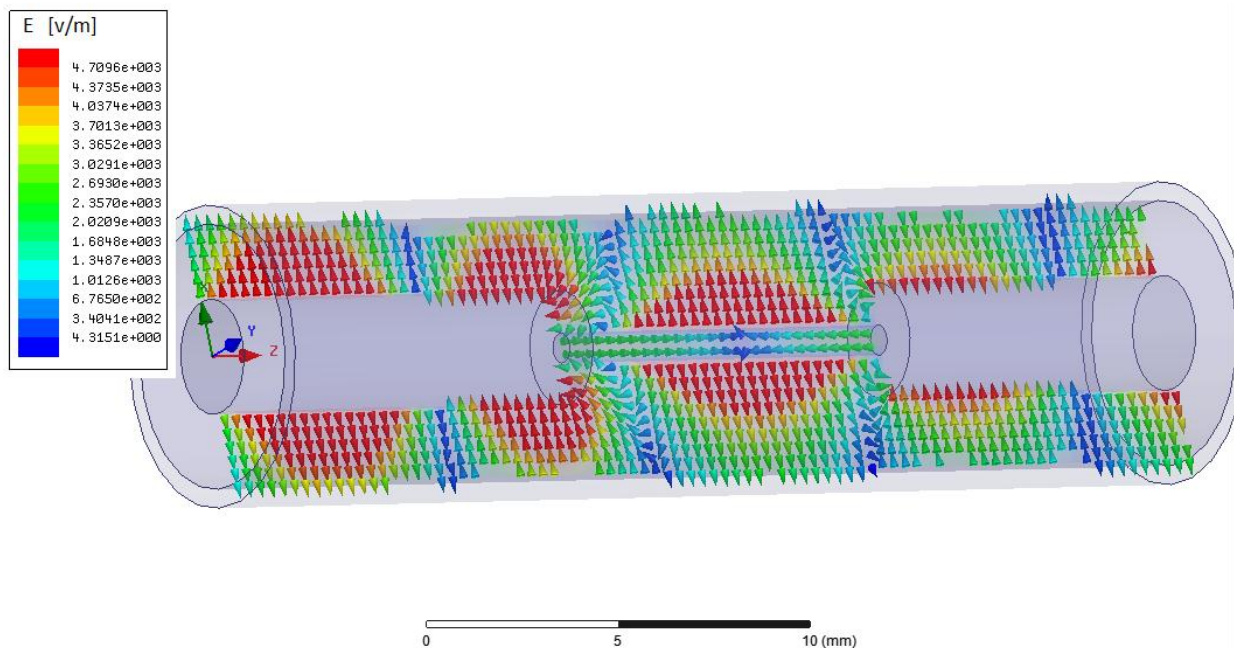


Fig. 10. Electric field distribution in coaxial line with the long sample on high frequency (20 GHz).

Multimode capacitor model was tested by software based on FEM methods on extreme conditions: the electric field in the sample was non-homogeneous, the width/length ratio was small, the length of the sample was big (half wavelength) as shown in fig. 10. Sample dimensions are  $R_s=0.4\text{mm}$   $d=10\text{mm}$ . Permittivity is  $\epsilon=200$ ,  $\tilde{\epsilon}=20$ .

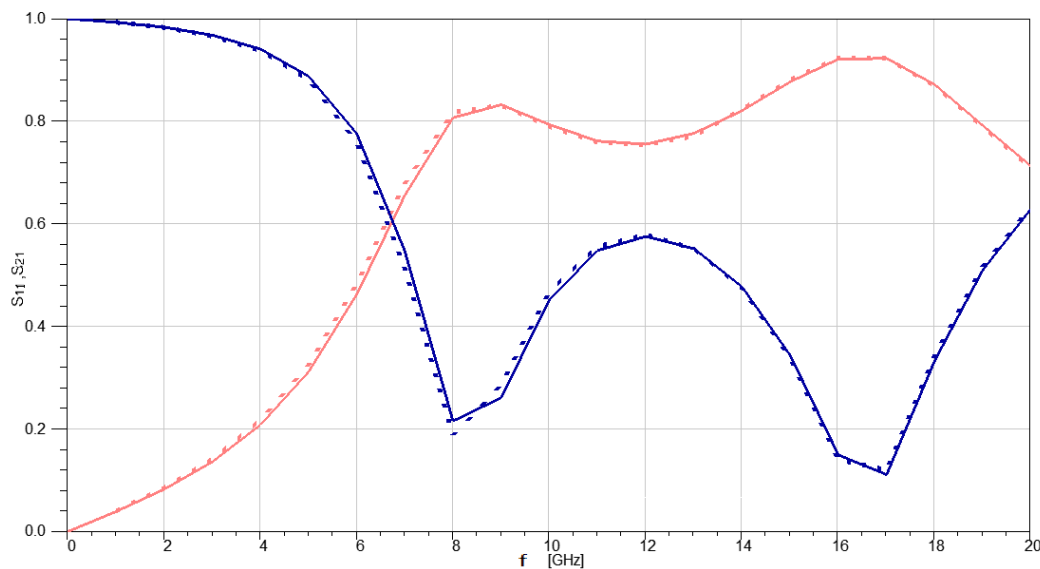


Fig. 11. Calculated magnitude of the reflection (blue) and transmission (red) coefficients. Solid lines represents FEM calculations, dotted lines - multimode capacitor method calculations.

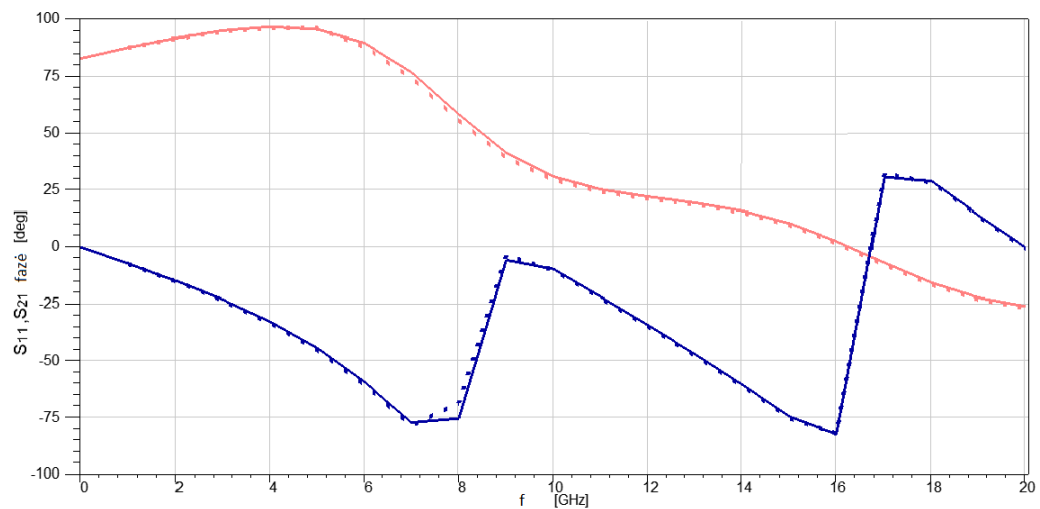


Fig. 12. Calculated phase of the reflection (blue) and transmission (red) coefficients. Solid lines represents FEM calculations, dotted lines- multimode capacitor method calculations.

Calculated by both FEM and multimode model S parameters shows excellent agreement (fig. 11 and fig. 12). The resonance on lower frequency appears due field distribution in

the sample radius. The second high frequency resonance appears, because long sample presents half wave transformer. Mention should be made, it is impossible to obtain second resonance using dynamic capacitor model. Unlike on low frequencies, there is strong magnetic field in the sample on high frequencies (fig. 13). The magnetic field in the sample is much stronger than in the transmission line. Thus, the magnetic permeability of the sample will affect to scattering parameters. There are S parameters of the sample like in fig.10. (while  $\mu = 1$  and  $\mu = 1.5$ ) presented in the table 1. Consider table data on frequency 8 GHz and 12 GHz (highlighted cells), it is possible to do magnetic permeability measurements on high frequency, using multimode capacitor model.

Freq. GHz	$S_{11}$				$S_{21}$			
	Magnitude		Phase		Magnitude		Phase	
	$\mu = 1$	$\mu = 1.5$	$\mu = 1$	$\mu = 1.5$	$\mu = 1$	$\mu = 1.5$	$\mu = 1$	$\mu = 1.5$
2	0.9829	0.9828	-15.04	-15.06	0.0855	0.0860	91.20	91.15
4	0.9383	0.9360	-33.14	-33.43	0.2159	0.2204	95.70	95.85
8	0.1921	0.1625	-69.94	-44.13	0.8168	0.8256	55.66	49.31
12	0.5790	0.5952	-34.77	-38.6	0.7552	0.7452	21.08	16.42

Table 1. S parameters dependence on magnetic permeability.

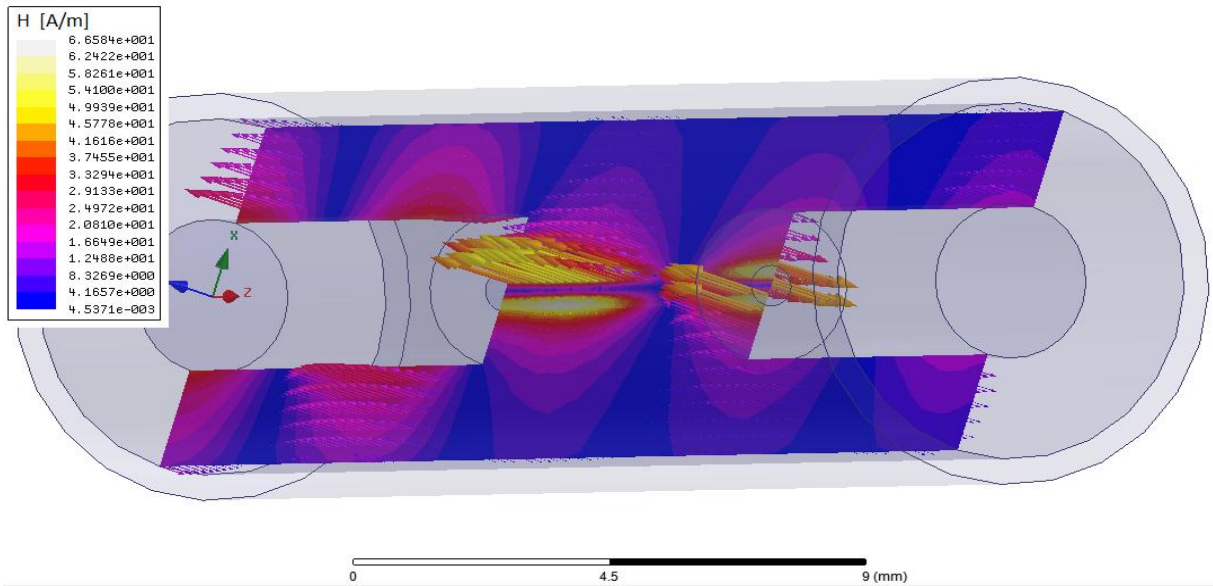
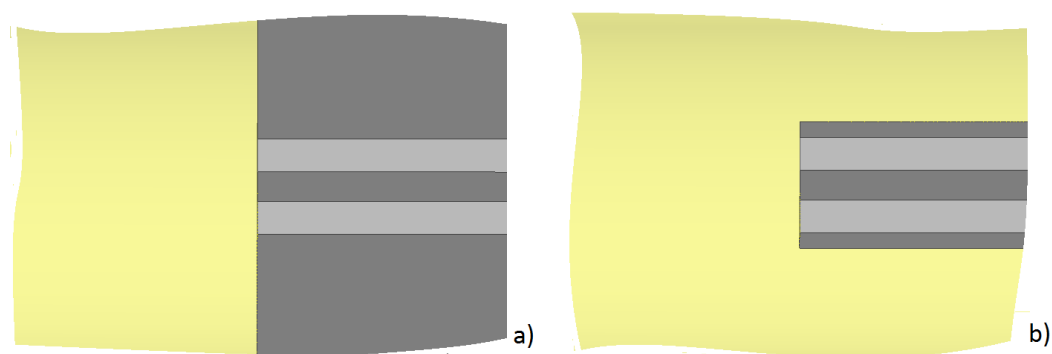


Fig. 13. Magnetic field distribution in coaxial line with the long sample on high frequency (20 GHz).



Possibility to measure low magnetic permeability when dielectric permittivity is high may be useful feature making both permittivity and permeability measurements of multiferroics or various ferrous composite materials.

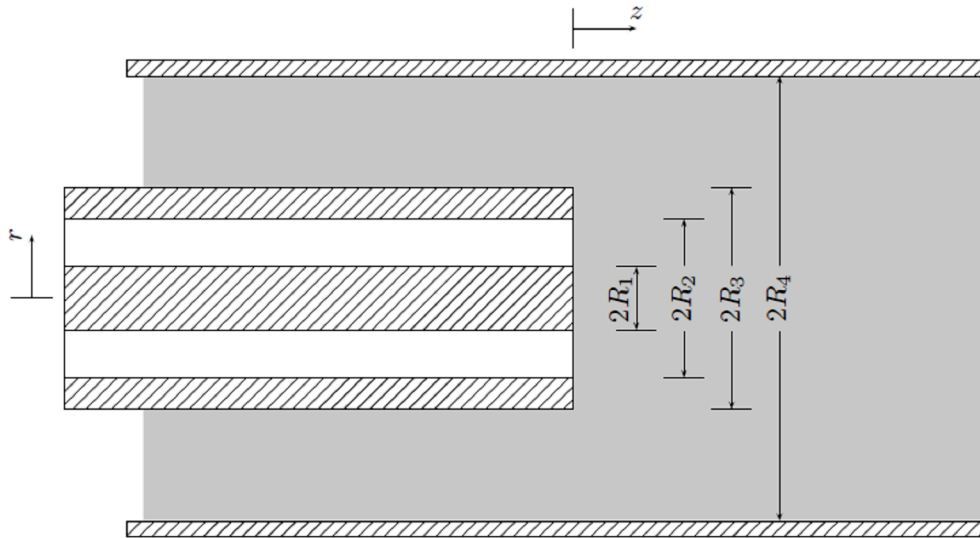
Measurement accuracy using capacitor models strongly depends on samples fabrication accuracy. To reduce sample preparation cost, another measurement methods like open ended coaxial line methods fig.14 may be chosen. Usually these methods are based on infinite sample assumption. Thus they are insensitive to sample's dimensions.



*Fig. 14. Types of conventional open ended coaxial lines.*

Real samples may have small dimensions wherefore infinite sample models may be not acceptable for dielectric permittivity calculations. Mathematical model and corresponding software for permittivity calculation was developed. Mathematical model was made on same multi mode approach like multimode capacitor model.

We consider the measurement setup (fig. 15) consists of three regions: a) coaxial line (probe), b) circular waveguide formed by the conducting walls of the container filled with the measured material, c) another coaxial line formed by the outer surface of the probe and the wall of the container filled with the measured material. The reflection factor is a result of scattering from the interface between the regions a) and b) which is also influenced by diffraction into the region c). It is assumed that dimensions of the coaxial line  $R_1$  and  $R_2$  are such, that only main (TEM) mode can propagate at the measuring frequencies. It is also assumed that the coaxial line is long enough and the container is deep enough so that the radiated electromagnetic waves do not reach neither the bottom of the container, or the surface of the material. Of course, this implies some degree of EM absorption in the material.



*Fig. 15. Open ended coaxial line with limited radius of sample.*

Multimode ended coaxial line model like multimode capacitor model was tested by HFSS software for the case described in fig. 16. Calculated by both FEM and multimode model reflection response in fig. 17 shows excellent agreement. Cases with increased radius and sample end metallization were calculated using HFSS software in order to estimate finite sample length and sample radius influence to scattering results. 3 methods of open end calibration and sample mounting device were suggested.

**Chapter 4** is devoted to lead-free ferroelectric ceramics with a perovskite structure based on  $(\text{Na}_{0.5}\text{Bi}_{0.5})\text{TiO}_3$  (NBT) and  $\text{La}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  (LMT) solid solutions investigation using broadband dielectrics spectroscopy methods. Dielectric properties of the NBT/LMT ceramics in 100 Hz  $\delta$  1 GHz frequency band were presented in [3].

In this work, the  $x\text{NBT}\delta(1-x)\text{LMT}$  ceramics with  $x$  equal to 0.80, 0.85, 0.90 and 0.95 were studied in 300  $\delta$  820 K temperature range and 1 MHz  $\delta$  1 GHz frequency band.

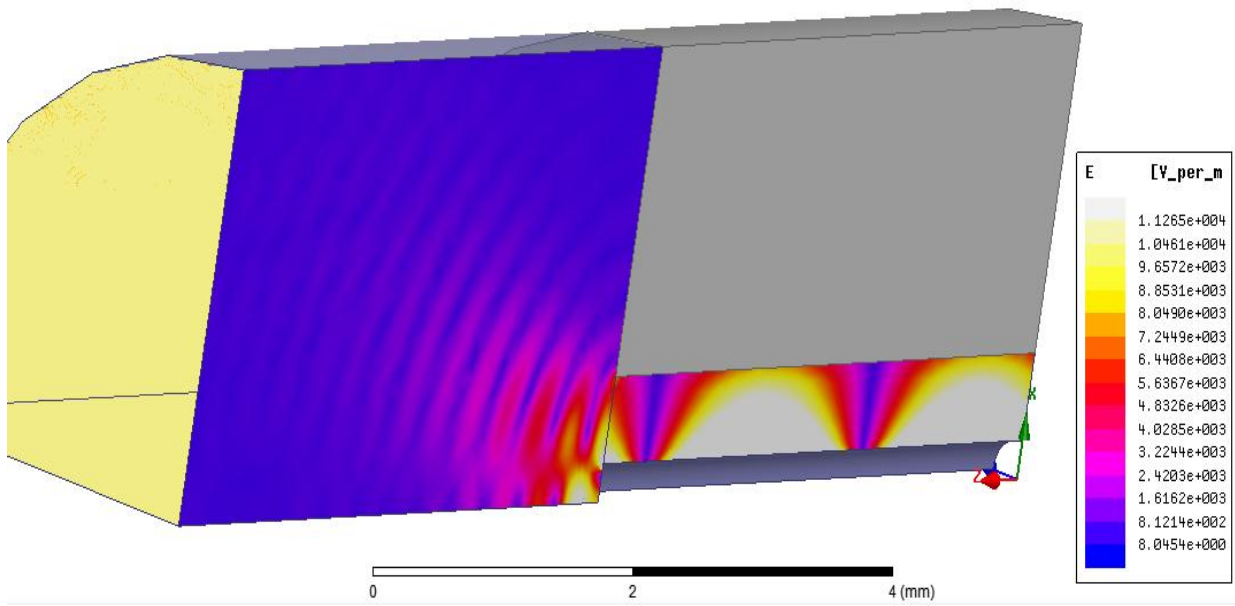


Fig. 16. Open ended coaxial line when  $R_3=R_4$ . Only quarter of measurement setup is shown.  $\text{Freq.}=40\text{ GHz}$   $R_1=0.255\text{mm}$ ,  $R_2=0.83\text{mm}$ ,  $R_3=R_4=3\text{mm}$ , dielectric permittivity of cable insulator - 2.1, permittivity of the sample -  $\epsilon=100$ ;  $\delta=10$ .

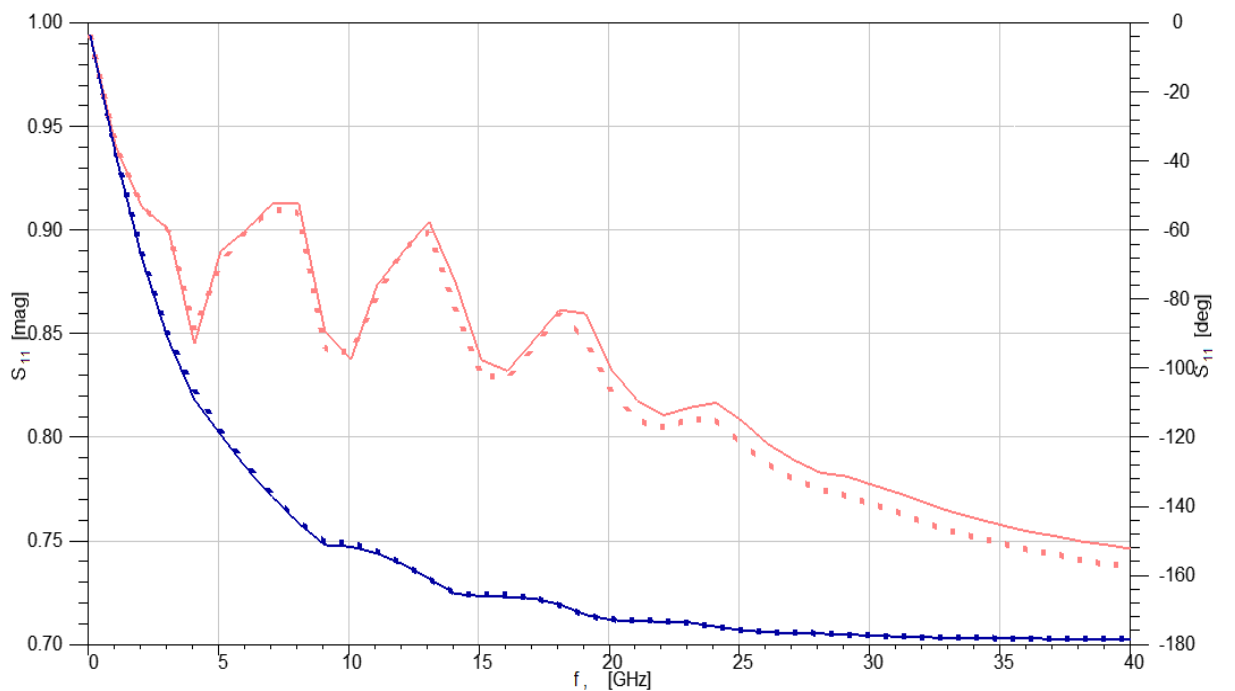


Fig. 17. Calculated magnitude (red) and phase (blue) of the reflection coefficient. Solid lines represent FEM calculations, dotted lines - multimode capacitor method calculations.

Dielectric measurements were carried out using a vector network analyzer Agilent 8714ET with a coaxial line setup. As coaxial line is highly prone to oxidation, the measurements were done in argon atmosphere on cooling with a rate of 1 K/min. All the samples were coated with Pt electrodes.

The impedance parameters were calculated using the dielectric data obtained from the coaxial line measurements.

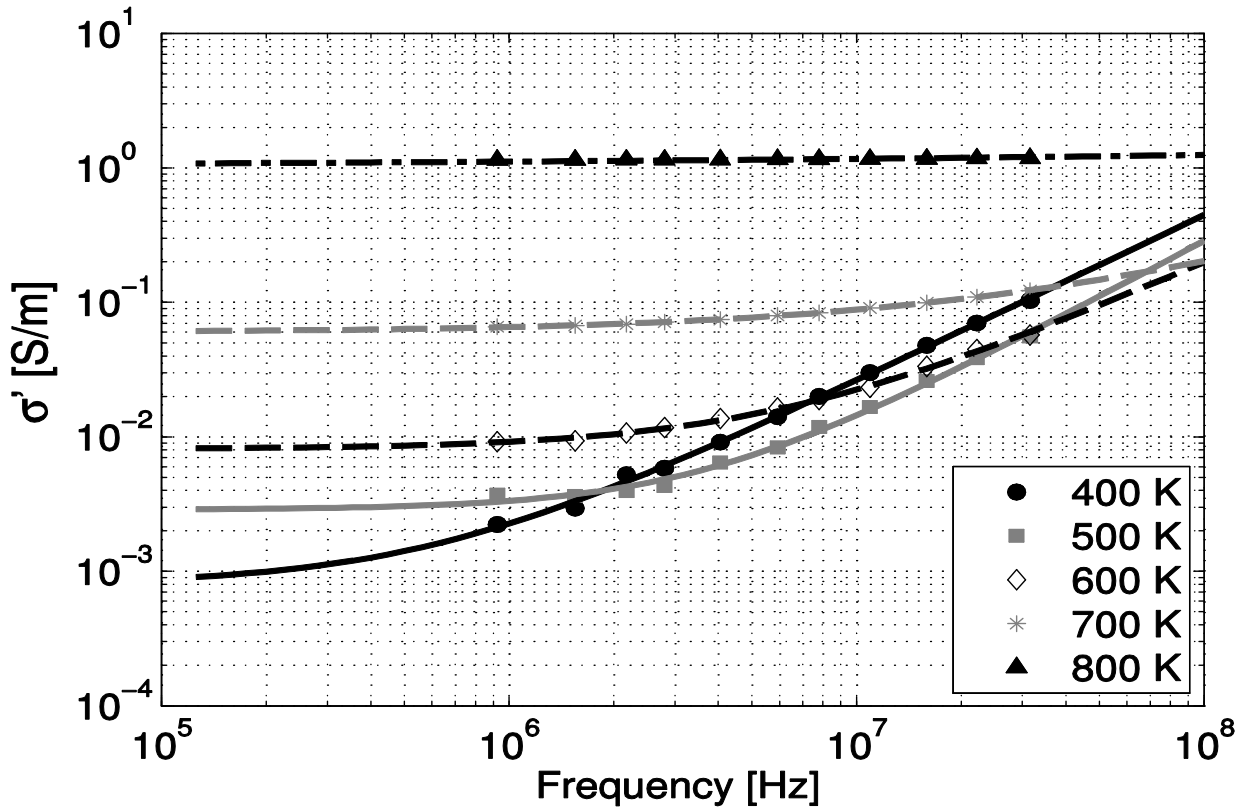


Figure 18. Frequency dependence of electric conductivity of  $x\text{NBT}\acute{o}(1-x)\text{LMT}$  ceramics with  $x = 0.95$ .

Very high conductive losses were observed due to the argon atmosphere. These losses can be associated with loss of oxygen leaving ceramics at such conditions, thus forming mobile vacancies. That is why the conductivity-temperature dependencies were calculated from the dielectric data. It is possible to derive DC conductivity value having sufficient frequency data.

Solid lines in fig. 18 represent the fit of the data with the following equation:

$$\sigma = A \cdot f^n + \sigma_{DC}$$

Here,  $A$  and  $n$  are constants which describe frequency dependence of conductivity, and  $\sigma_{DC}$  is DC conductivity. In all cases the frequency range of 1 MHz  $\delta$  40 MHz was used to fit experimental data.

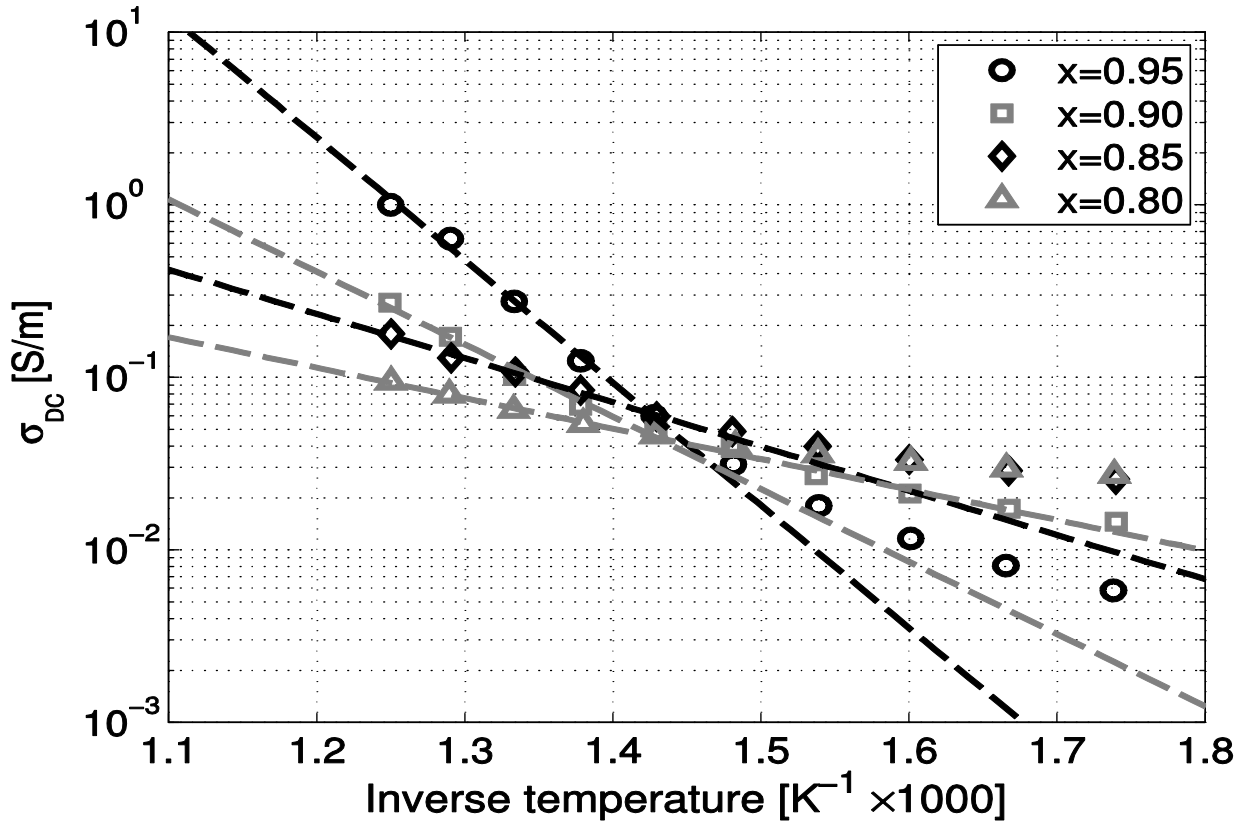


Figure 19. Static conductivity of  $x\text{NBT}\delta(1-x)\text{LMT}$  ceramics with different values of mole fraction of NBT as a function of inverse temperature.

Dependencies of static conductivity for all ceramics on inverse temperature are shown in figure 19. Solid lines represent the Arrhenius law fit:

$$\sigma_{DC} = \sigma_{DC0} \cdot e^{-\frac{E_A}{kT}}$$

Here,  $\sigma_{DC0}$  represents infinite temperature static conductivity,  $E$   $\delta$  activation energy,  $T$   $\delta$  temperature and  $k$   $\delta$  Boltzmann constant.

Step changing of activation energy was obtained on 690K, which is probably related to phase transition like in pure NBT from paraelastic to ferroelastic phases [4]. The activation energy and  $\sigma_{DC0}$  dependence vs. NBT concentration is shown on fig. 20.

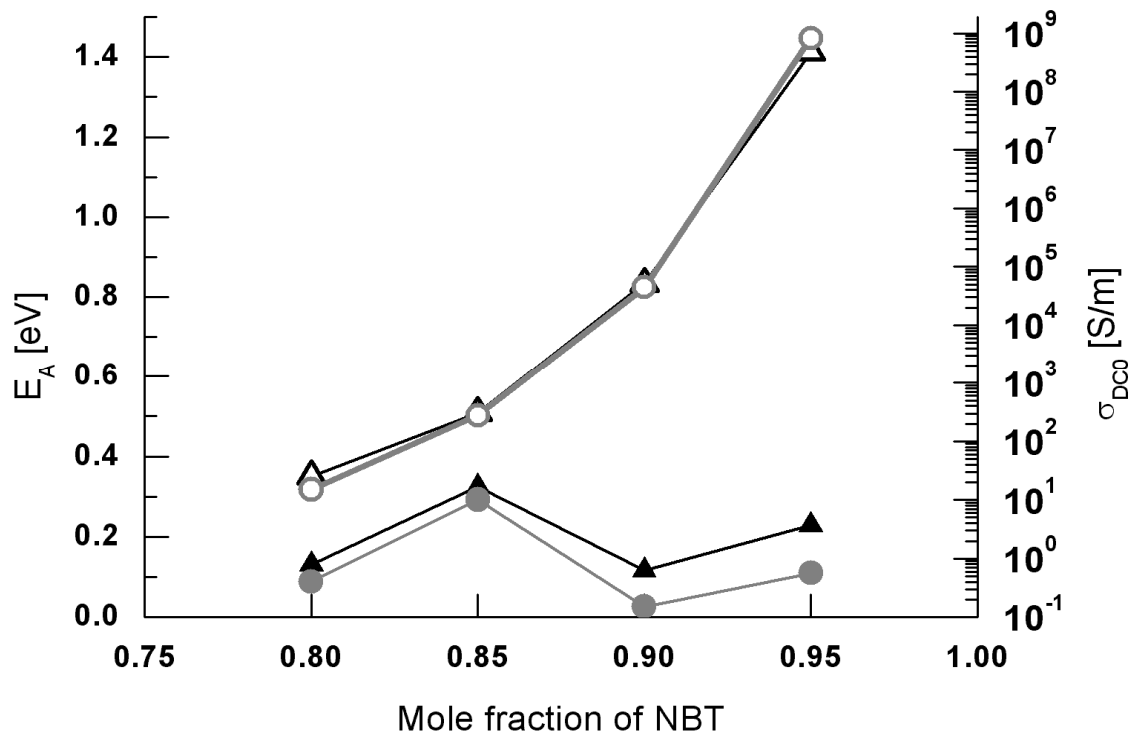


Figure 20. Activation energy (triangles) and conductivity prefactor (circles) of the static conductivity of  $x\text{NBT}\delta(1-x)\text{LMT}$  ceramics versus the NBT mole fraction at the temperatures lower 690K (filled dots) and higher 690K (empty dots).

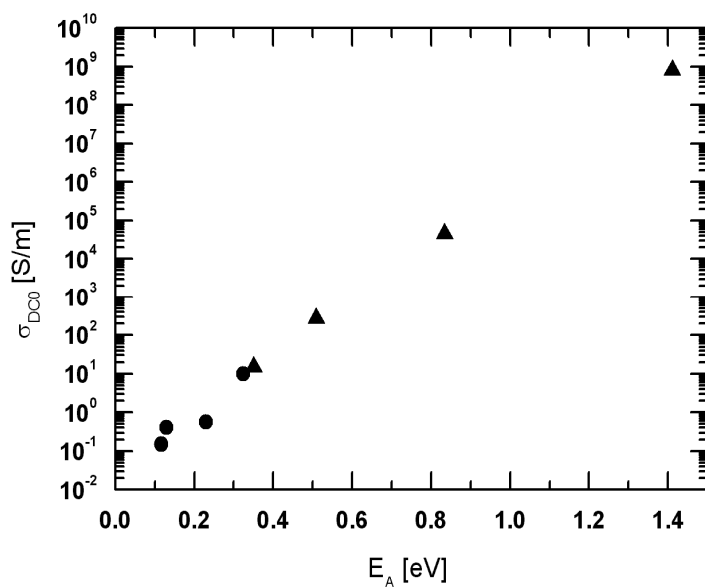


Figure 21. MN plot at the temperatures lower 690K (circles) and higher 690K (triangles).

It looks like  $E_A$  dependence on  $\log(\sigma_{DC0})$  is linear:

$$\log(\sigma_{DC0}) = a + b \cdot E_A$$

Here  $a$  and  $b$  are constants. This relation is known as the Meyer-Neldel (MN) rule [5].

The performed measurements allow to conclude that increase of LMT content lowers the activation energy of static electric conductivity, which is related to oxygen vacancies. At high temperatures (above 700 K) conductivity is lower when LMT content is increased.

At fig. 21 we can see a straight line, which means that MN rule is valid at both phases.

The origin of MN rule in NBT6LMT ceramics is not known yet.

## Conclusions

1. Using optimization option in the commercial EM simulation software it is possible to find the electrical material properties of the samples in very complex shape. Usage of numerical methods allows us to take into account the spurious factors arising at connection of a measured sample to a metering circuit. Thus, it is possible to consider the EM simulation software as a powerful and universal tool for the material properties measurements.
2. Multimode capacitor allows to calculate values of dielectric permittivity much more accurately, especially in the high frequency range, avoiding very time consuming FEM or any other numerical method. At the high frequencies (or when permittivity is high) when distribution of the electric field in the sample is non-homogeneous, it is possible to measure both dielectric permittivity and magnetic permeability using multimode capacitor model.
3. Modified open ended coaxial model is suitable for dielectric permittivity measurements of various dimension samples.
4. Transition from paraelastic to ferroelastic phase temperature for the investigated NBT-LMT ceramics group does not depend on LMT concentration.
5. Meyer-Neldel rule for electric conductivity is valid in NBT-LMT ceramics group.



## References

- [1] Rudys S, Ivanov M, Banys J: EM simulation for the material electrical properties measurement in microwave range. Proceedings of the 16th international student seminar "Microwave and optical applications of novel phenomena and technologies", June 89, Oulu, Finland. 73 (2009).
- [2] Roberts, S.= Von Hippel, A: A New Method for Measuring Dielectric Constant and Loss in the Range of Centimeter Waves. J. Appl. Phys. 17: 610-616 (1946).
- [3] Saulius Rudys, Maksim Ivanov, Juras Banys, Nikolai P. Vyshatko, Aleksandr D. Shilin & Andrei N. Salak: Dielectric and Impedance Spectroscopy of  $x\text{NBT}(1-x)\text{LMT}$  Ceramics, *Ferroelectrics*, 417:1, 143-150 (2011).
- [4] Dorcet, V; Trolliard, G.; Reinvestigation of Phase Transitions in  $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$  by TEM. Part II: Second Order Orthorhombic to Tetragonal Phase Transition, *Chemistry of materials* 20 (15) (2008)
- [5] W. Meyr and H. Neldel, *Z. Techn. Phys. (Leipzig)* 12, 588 (1937).

## List of publications

1. Ivanov Maksim, Banys J ras, Rudys Saulius, Grigalaitis Robertas. Measurements of complex dielectric constant of ferroelectrics with six-port reflectometer in 80-120 GHz frequency range // *Ferroelectrics*. 2008, vol. 367, p. 229-233 <http://dx.doi.org/10.1080/00150190802377603>
2. Banys J ras, Rudys Saulius, Ivanov Maksim, Li Jing, Wang Hong. Dielectric properties of cubic bismuth based pyrochlores containing lithium and fluorine // *Journal of the European Ceramic Society*. 2010, vol. 30, no. 2, p. 385-388. <http://ecers.org/pages/publications/journal.phpvol.30%20n.2%20pp:385-388%20>
3. Rudys Saulius, Ivanov Maksim, Banys J ras, Vyshatko Nikolai P. , Shilin Aleksandr D. , Salak Andrei N. Dielectric and Impedance Spectroscopy of xNBT-(1-x)LMT Ceramics // *Ferroelectrics*. 2011, vol. 417, iss. 1, p. 143-150. <http://dx.doi.org/10.1080/00150193.2011.578530>
4. Banys J ras, Lapinskas Saulius, Rudys Saulius, Grei ius Simonas, Grigalaitis Robertas. High frequency measurements of ferroelectrics and related materials in coaxial line // *Ferroelectrics*. 2011, vol. 414, iss. 1, p. 64-69. <http://dx.doi.org/10.1080/00150193.2011.577308>
5. S. Rudys, M. Ivanov, J.Banys. Ansoft HFSS software application for the dielectric and magnetic measurements of ferroelectrics and related materials in microwaves // *Ferroelectrics* 2012, vol. 430, iss. 1, p. 115-122. <http://www.tandfonline.com/doi/abs/10.1080/00150193.2012.677732>
6. J. Banys , J. Grigas , S. Rudys , I. Kosakovskis . Investigation of TiGaSe<sub>2</sub> crystal dielectric properties in frequency range 80-120GHz//, *Lithuanian J. of Physics* . 1992 ,vol. 32, nr. 2 , p. 260-266.
7. Rudys Saulius, Ivanov Maksim, Banys J ras. EM simulation for the material electrical properties measurement in microwave range // *Proceedings of the 16th international student seminar "Microwave and optical applications of novel phenomena and technologies"*, June 8-9, Oulu, Finland. Oulu, 2009, p.73. <http://herkules.oulu.fi/issn03553213/index.html?ang=en>

8. Ivanov Maksim, Rudys Saulius, Lapinskas Saulius, Banyss J ras, Macutkevi Jan, Yermakov A. Ye. , Uimin M. A. , Mysik A. A. , Shenderova Olga. Measurements of complex dielectric permittivity and magnetic permeability of carbon-coated Ni capsules // MIKON 2010: 18th international conference on microwaves, radar and wireless communications: [proceedings], ISBN 978-9955-690-19-1 Vilnius, June 14-16, 2010. Vol. 1. Vilnius: Geozondas, 2010, p. 105-109.  
<http://ieeexplore.ieee.org/Xplore/login.jsp?url=http%3A%2F%2Fieeexplore.ieee.org%2Fiel5%2F5523194%2F5540324%2F05540424.pdf%3Farnumber%3D5540424&authDecision=-203>

## **About the author**

Saulius Rudys was born on May 2, 1965 near Ignalina. During the period of 1972-1983 he studied at Jonava 4<sup>th</sup> secondary school. In 1983 he entered the Faculty of Physics of Vilnius University. In 1990 graduated it. During 1984-1986 served in soviet army. During the period of 2007-2011 Saulius Rudys studied in a doctoral program of Physics at Vilnius University.

During 1990-1993 Saulius Rudys worked at Vilnius at Vilnius Scientific Research Institute of Radio-Measurement Instruments as engineer. Since 1993 he works at UAB „Elmika“ as director of antennas and passive components department. Since 2011 Saulius Rudys works at Vilnius University as engineer and at State scientific research institute Center for Physical Sciences and Technology as junior researcher. Main area of interest of Saulius Rudys is research and development of antenna systems, millimetre wave components and measurement equipment.

Since doctoral studies Saulius Rudys regularly presents his research results in various international conferences. He has published 8 scientific publication in peer reviewed Lithuanian and international periodical, all of them concern a topic of a dissertation, 5 publications are in the journals with ISI citation index.

Saulius Rudys is married; his wife Violeta is economist. He has two children.

## Santrauka

Šiame darbe nagrinėjamos galimybės patobulinti plačiajuosčių dielektrinės spektroskopijos metodus naudojant skaitmeninius ir analitinius daugelio mod dielektrinės skvarbos skaičiavimo metodus, tiriami  $(1-x)(\text{Na}_{1/2}\text{Bi}_{1/2})\text{TiO}_3 - x\text{La}(\text{Mg}_{1/2}\text{Ti}_{1/2})\text{O}_3$  (NBT-LMT) keramikų grupės laidumo ypatumai.

Disertacijos vade yra aptariama tyrimo sritis ir aktualumas, pristatomas tyrimo objektas, formuluojami ginamieji teiginiai, pristatomas mokslinis naujumas, publikacijų disertacijos tema straipsniai, konferencijose, kuriose buvo pristatyti rezultatai disertacijos tema, straipsniai ir disertacijos turinys.

Pirmame skyriuje pristatoma tarptautinės mokslinės literatūros apžvalga, liečianti dielektrinės skvarbos matavimo metodikas, kiek plačiau apžvelgiant atviro galo koaksialines linijas metodus.

Antrame skyriuje pristatoma tarptautinės mokslinės literatūros apžvalga, liečianti NBT-LMT ir giminingų medžiagų savybes.

Trečiame skyriuje nagrinėjama galimybė pritaikyti HFSS skaitmeninio modeliavimo programą dielektrinės spektroskopijos tikslams. Naudojant šią programą, apskaičiuojama dielektrinė ir magnetinė skvarbos komplikuotiems mikrojuostelinėms linijoms ir dalinai užpildyto bangolaidžio matavimo grandinėms atvejais. Pateikiami patobulinti kondensatoriaus koaksialinėje linijoje ir ribotai matmenų atviro galo koaksialinėms linijoms matematiniai modeliai. Šie modeliai patikrinami skaitmeniniu metodu. Naudojant daugiamodulį kondensatoriaus modelį, atsižvelgus magnetinio lauko pasiskirstymą koaksialinėje matavimo grandinėje, pasiūlymas bandomas pamatuoti mažai bandinio magnetinei skvarbai, kai dielektrinė skvarba didelė (dešimt ar daugiau kartų didesnė už magnetinę skvarbą). Atviro galo koaksialinei linijai siūloma keletas kalibravimo būdų, reikšmingi bandiniui prispausti prie linijos.

Ketvirtame skyriuje nagrinėjama bežvaidžių NBT-LMT keramikos dielektrinių savybių matavimo rezultatai. Gauti rezultatai rodo, kad NBT-LMT keramikų grupėje elektriniam laidumui galioja Maerio-Neldelio taisyklė.

Gautos išvados pateikiamos trečiame ir ketvirtame skyriuje.

Cituotos literatūros straipsniai pateikiamas kiekvieno skyriaus gale.

## Apie autori

Saulius Rudys gim 1965 m. Gegufl s 2 d. Ignalinos rajone. Nuo 1972 m. Mok si Jonavos 1-ojoje vidurin je mokykloje, v liau 4-ojoje vidurin je mokykloje (dabar Jonavos Senamies io gimnazija), kuri 1983 m. baig . 1983-1990 m. su dviej met tarnybos armijoje pertrauka, studijavo Vilniaus universiteto Fizikos fakultete. 2007-2011 m. studijavo Vilniaus universiteto fizini moksl krypties doktorant roje.

1990 ó 1993 m. Saulius Rudys dirbo inffinieriumi Vilniaus radijo matavimo prietais institute. Nuo 1993 m. iki -iol dirba UAB šElmikaõ Anten ir pasyvini komponent skyriaus vir-ininku, direktoriaus pavaduotoju. Nuo 2011m. iki -iol dirba jaunesniuaju mokslo darbuotoju Valstybinio mokslini tyrim instituto Fizini ir technologijos moksl centre (projektas šEkranuojan i kompozicini medfliag su anglies nano vamzdeliais tyrimas ir taikymasõ) ir inffinieriumi Vilniaus universiteto Fizikos fakulteto radiofizikos katedroje. Sauliaus Rudffio pagrindin profesini interes sritis ó vairi anten sistem k rimas, superauk-to daffnio matavimo prietais ir j auk-tadaffni mazg k rimas.

Saulius Rudys nuo doktorant ros pradffios nuolat dalyvauja ir pristato darbo rezultatus vairiose tarptautin se konferencijose. Jis yra i-spausdin s disertacijos tema 8 publikacijas, 5 j - ISI s ra-o flurnale.

Saulius Rudys yra ved s, flmona Violeta - ekonomist . Turi du vaikus.