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Intercomparison of permittivity measurement techniques for ferroelectric thin layers

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The dielectric properties of a $\text{KTa}_{0.65}\text{Nb}_{0.35}\text{O}_3$ ferroelectric composition for a submicronic thin layer were measured in the microwave domain using different electromagnetic characterization methods. Complementary experimental techniques (broadband methods versus resonant techniques, waveguide versus transmission line) and complementary data processing procedures (quasi-static theoretical approaches versus full-wave analysis) were selected to investigate the best way to characterize ferroelectric thin films. The measured data obtained from the cylindrical resonant cavity method, the experimental method that showed the least sources of uncertainty, were taken as reference values for comparisons with results obtained using broadband techniques. The error analysis on the methods used is discussed with regard to the respective domains of validity for each method; this enabled us to identify the best experimental approach for obtaining an accurate determination of the microwave dielectric properties of ferroelectric thin layers.

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I. INTRODUCTION

Electromagnetic (EM) characterization of materials is a prerequisite step before their use in microwave applications: radar, mobile phones, wireless communications, etc. It enables circuit designers to precisely determine of the EM properties (permittivity, permeability, and electric conductivity) of the materials composing studied devices or the systems. It also allows quality control of materials in the manufacturing process and validation of models of interaction between EM waves and matter by comparison with experiments. It is widely recognized that poor design of microwave devices is often caused by erroneous values of the EM properties of the circuit substrate.

Many measurement techniques have been developed over recent years to determine the EM properties of materials. These are based on the measurement of the perturbation of propagating or standing waves (e.g., coaxial line-based reflection/transmission method, cavity perturbation technique, etc.). Commercialization of high-performance vector network analyzers (VNA) has led to a gradual increase in the automation of these techniques.

Today, the characterization of homogeneous and isotropic dielectric thick samples is conducted relatively well using conventional methods with transmission lines or resonant cavities, combined with suitable data processing programs. However, this is not the case for materials that exhibit an unusual shape, such as thin films and painted layers; in such cases, obtaining an accurate measurement is still problematic. Difficulties are increased when studying

the influence of external parameters (such as a mechanical stress, shift in temperature, or DC biasing field) on the EM properties of these materials.

Ferroelectric thin films fall into this category of materials. Their submicron thickness introduces the problem of the sensitivity of the measurement method used. The study of variations in their dielectric properties under the action of a static electric field requires specific techniques with command electrodes. Finally, the influence of the metallizations, caused by possible inter-diffusion, and of the type of substrate chosen for thin layer deposition (mono- or polycrystalline), must also be examined.

Solving the problem of how to perform such an analysis is paramount because these materials are needed to fit the specifications of new applications in different domains. Thin films are considered very promising materials for improving RF circuit performances through new functionalities: tunability and miniaturization of filters, phase shifters, antennas, etc.

In this context, the French laboratories XLIM, IMEP-LAHC, and Lab-STICC, who are specialized in the EM characterization of materials, decided to conduct an intercomparison of permittivity measurements for ferroelectric thin films. The strategy adopted was to use different characterization techniques to measure the complex permittivity of a single ferroelectric composition $\text{KTa}_{0.65}\text{Nb}_{0.35}\text{O}_3$ (KTN) deposited on different substrates at the ISCR laboratory. Even though the selected methods do not necessarily operate at the same frequencies because of their own domain of validity, all experimental data collected were obtained in the

centimeter wave range (3–30 GHz) of the microwave domain. Each laboratory involved in the intercomparison procedure conducted its own characterization without knowing the results obtained by the other partners. The potential differences in temperature and humidity encountered in different laboratories during the measurements were not taken into account in this intercomparison experiment.

II. EM CHARACTERIZATION OF DIELECTRIC THIN FILMS

Permittivity is a physical quantity that describes the ability of a medium to become polarized (appearance of dipolar moments) under the action of an electric field. In the dynamic regime, in order to account for dissipative effects in the matter, ε is represented by a complex quantity ($\varepsilon = \varepsilon' - j\varepsilon''$) that depends on the field frequency (dispersion), the homogeneity of the medium, its possible anisotropic character and external parameters such as temperature and mechanical stresses.¹ In the case of ferroelectric media, permittivity varies if the polarization state of the material is changed by the application of a static electric field: a property used to ensure the tunability of microwave functions (filters, phase shifters, etc.).^{2–7} This offers an interesting alternative to the more conventional technological solution based on the use of active elements (PIN diodes, varactors) and, more recently, micro-electromechanical systems (MEMS).

The EM characterization of a ferroelectric thin layer consists in determining, at one frequency or over a wide range, its relative permittivity compared with a vacuum $\varepsilon_r = \varepsilon/\varepsilon_0$ and its loss tangent $\tan\delta = \varepsilon''/\varepsilon'$. Two operating procedures exist to measure these two physical quantities in the microwave domain. Mono-frequency methods use resonating modes in cavities.^{8–13} The insertion of a sample into the cavity induces a shift in its resonant frequency and quality factor. The measurement of these variations enables highly accurate determination of the quantities ε_r and $\tan\delta$ at a single frequency. Even if mono-frequency methods give very accurate results, broadband methods can also be used in the microwave spectrum since they allow the dispersion of EM properties of the constructed ferroelectric materials to be studied. To determine dielectric properties of materials over a wide range of frequencies, reflection/transmission measurement techniques, based on the use of rectangular waveguides or transmission lines such as the coaxial type, were developed in the late 20th century.^{14–16} These techniques have been successfully applied to characterize dielectric or magnetic homogeneous, isotropic materials in bulk form (thick samples). Intercomparison of measurements, carried out using the two major operating procedures (mono-frequency¹⁷ and broadband¹⁸), accompanied by an analysis of the different error sources affecting the measurements, made it possible to define the best experimental protocols and implementation of the most robust experimental data processing programs to ensure an accurate EM characterization of bulk materials. In this field, the reflection/transmission technique is based on the standard use of a coaxial line.¹⁹ Unfortunately, this regular method, which consists in inserting the sample to be characterized (torus of a few millimeters

thick) into the line, is poorly adapted for the characterization of thin films. Alternative solutions in planar technologies (microstrip or coplanar) started to appear in the 1990s to work around this inadequacy, whilst retaining the advantage of the broad operating band of transmission lines.^{20–22} The philosophy of this approach is to choose measurement cells that guarantee a strong interaction between the EM wave and the thin film, which is made possible by a favorable direction and concentration of the EM field with regard to the planar geometry of the material.

For ferroelectric thin films, the approach consists in integrating the layer, and its substrate, directly into the transmission line.²³ The metallic layers (strip and ground plane) are deposited on the thin layer, which is itself deposited on its own substrate. The measurement of the line propagation constant makes it possible to determine the complex relative permittivity of the material to be characterized.²⁴ Coplanar waveguide (CPW) structures seem to have the best suited topology for ferroelectric thin films because they ensure a tangential direction of the electric field compared with the plane of the layer, which is a required condition for good method sensitivity. CPW technology also allows easy integration of a static electrical command to study the DC field-dependent permittivity of the material. Another advantage of this solution relates to the *in-situ* character of the conducted measurement, because the permittivity of the layer is determined in a very similar configuration to those used in practice in CPW microwave devices. This is particularly important in the case of an anisotropic medium for which the values of the in-plane and out-of-plane permittivities are different.

However, broadband CPW methods are destructive, and the parameter determined from the measured scattering parameters (S-parameters) of the line corresponds in reality to the effective permittivity of the line and not that of the thin layer, which is only a part (with a very small volume) of the overall structure. To retrieve the complex relative permittivity of the thin layer, a final step must be implemented in order to reverse the electromagnetic analysis of the CPW structure. However, as the thin layer is integrated under the metallic layers of the CPW structure, it is impossible to perform a differential measure that would eliminate cell imperfections (metallic losses, radiation, dielectric losses of the thick substrate) from the measured S-parameters. Many error sources therefore affect the accuracy of the results. These are caused by uncertainties during the data processing procedure in certain geometric parameters (thickness of the thin layer or metallic layers) and electromagnetic parameters (permittivity of the substrate layer, electrical conductivity of the metallic layers,²⁵ or pollution that the latter can cause by inter-diffusion, radiation, or dispersion) of the line. In some cases, these different elements may finally have a greater influence on the measured parameters (S-parameters) than that generated by the ferroelectric thin film, which displays a very small thickness (typically, a few hundred nanometers).

The objective of the present intercomparison measurements is to identify the potential of different EM characterization methods for the accurate determination of the dielectric properties of ferroelectric thin layers. From these comparisons performed on a same ferroelectric thin film,

analysis of the obtained results should allow us, first, to define an experimental protocol and a data processing procedure for each method, to improve the accuracy of the results obtained and, ultimately, to define an EM characterization standard for dielectric thin films based either on a single method or on a combination of complementary methods in terms of accuracy and bandwidth.

III. KTN FERROELECTRIC THIN FILMS: SYNTHESIS AND MICROSTRUCTURAL PROPERTIES

Previous reports have investigated dielectric properties of $\text{KTa}_{0.6}\text{Nb}_{0.4}\text{O}_3$ thin films on polycrystalline alumina substrates in the microwave frequency range,^{26,27} but comparisons of the measured permittivity values would be risky because deposition processes differed: pulsed laser deposition²⁶ versus chemical solution deposition.²⁷ KTN thin films were deposited on various substrates suitable for the microwave range, especially LaAlO_3 and MgO .²⁸ In this study, 400 nm thick KTN films were deposited by pulsed laser ablation (KrF laser, $\lambda = 248$ nm) at 700 °C under 0.3 mbar oxygen pressure. This KTN composition was selected for its Curie temperature, T_c , which is slightly lower than room temperature (on bulk material $T_c = 269$ K²⁹), in order to favour a high tunability, while avoiding hysteretic effects at room temperature. On both LaAlO_3 and MgO substrates, the films exhibited a (100) epitaxial growth, as evidenced by X-Ray Diffraction performed in θ - 2θ , ω -, and φ -scan modes.²⁸ The structural quality appears to be higher on LaAlO_3 on which the full widths at half maximum, $\Delta\omega$ and $\Delta\varphi$, of the ω -scan (rocking-curve) and φ -scan are generally lower than on MgO .²⁸ On MgO , the presence of a secondary minor (110) orientation is commonly observed, as shown by Fig. 1. The Scanning Electron Microscopy (SEM) micrographs are in agreement with the XRD measurements, where (100) epitaxial thin films were revealed (in-plane ordering of plate square grains) on both substrates, in addition to the coexistence of the (110) secondary orientation on MgO substrate illustrated by the elongated grains) (Fig. 2). Despite the difference in crystalline quality, a KTN sample deposited on MgO was next chosen for the inter-comparison of microwave measurements. The main reason for this choice is the lower permittivity of MgO ($\epsilon_r \sim 9$ -10), which is isotropic, whereas the presence of twins in LaAlO_3 can induce modifications in its permittivity, which is higher ($\epsilon_r \sim 24$) than that of MgO . A value that is too high for the substrate permittivity would induce a decrease of the sensitivity of the measurement method used for the EM characterization of the ferroelectric layer.

IV. DESCRIPTION AND PRINCIPLES OF MICROWAVE CHARACTERIZATION METHODS

This section will briefly present the experimental principle of the EM characterization methods used by the different laboratories in this intercomparison.

A. Rectangular waveguide method

The first method, developed at Lab-STICC, is based on the measurement of the scattering parameters (S-parameters)

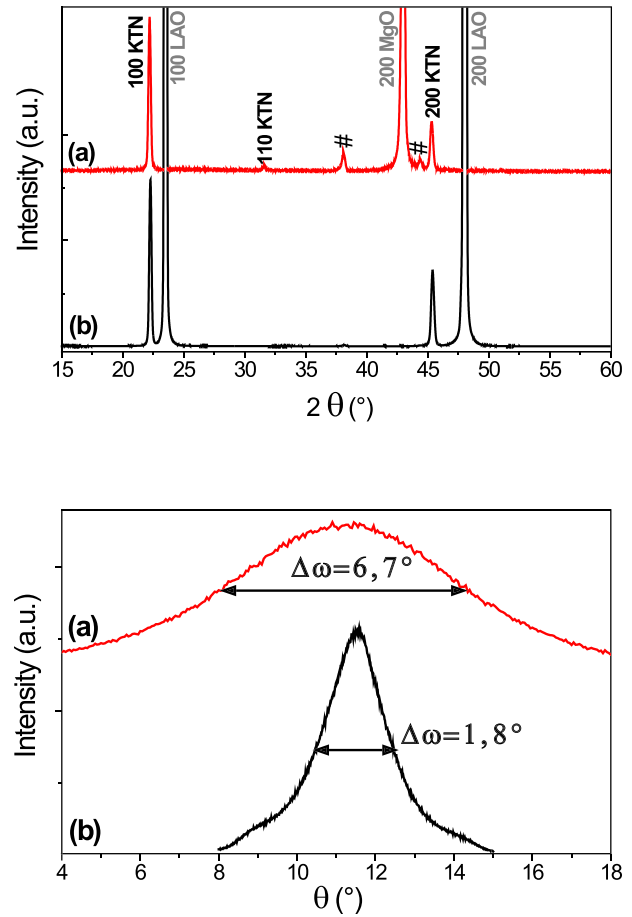


FIG. 1. θ - 2θ XRD patterns of the KTN (65/35) grown on (100) MgO (a) and (100) LaAlO_3 (b) substrates. # indicates diffraction peaks of the sample holder. The (100) orientation of LaAlO_3 refers to the pseudo cubic lattice.

of a rectangular waveguide loaded by the ferroelectric thin film deposited on its own substrate.³⁰ As illustrated in Fig. 3, the square-shaped bilayer sample under study is set on its thickness in the mid-plane of the cross-section of an X-band (8–12 GHz) rectangular waveguide. A TRL calibration procedure enabled us to correct systematic errors and to shift the reference planes closer to the sample to be characterized.

Dynamic analysis of the measurement cell (mode-matching method) consists in evaluating the electric- and magnetic-field components that are solutions of the Maxwell equations; this enables the calculation of the direct problem and determination of the theoretical S-parameters of the loaded waveguide. This approach allowed us to take into

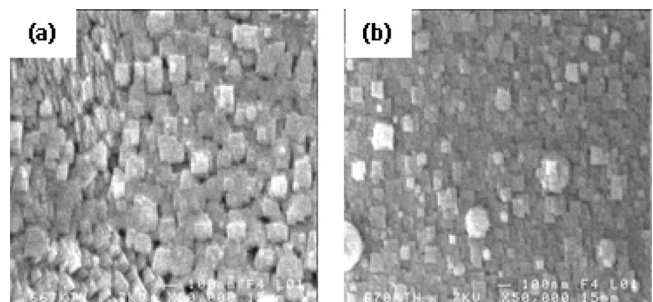


FIG. 2. SEM micrographs of the KTN thin films deposited on (a) MgO and (b) LaAlO_3 .

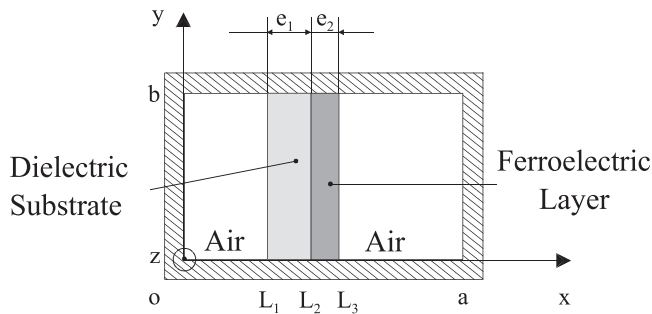
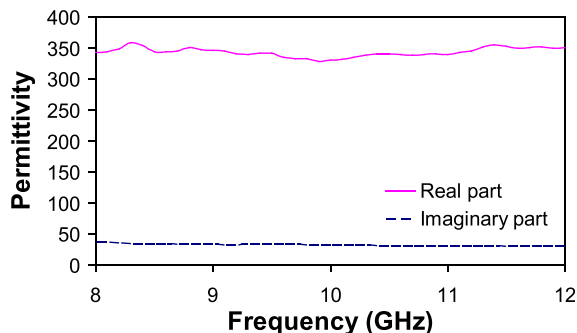


FIG. 3. Cross-section of the rectangular waveguide partially filled with a thin ferroelectric layer deposited on its substrate.

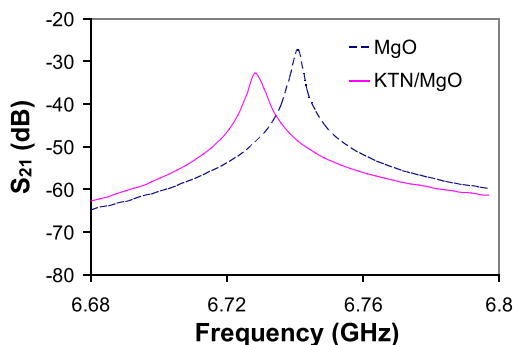
account not only the dominant mode (TE_{10}), but also the higher order modes that can be excited at the discontinuities in the waveguide.³¹

The waveguide cross-section was separated into 4 layers (air/dielectric/thin film/air). The dispersion relationships for each mode were established. A mode-matching analysis was then used to take into account the abrupt discontinuities between the empty waveguide regions and the one partly filled with the ferroelectric layer being characterized. Finally, the complex dielectric constant was extracted by minimizing the quadratic error between the measured S-parameters and those issued from the direct problem.

Fig. 4(a) shows the permittivity spectrum of a KTN thin film deposited on MgO substrate, extracted using the rectangular waveguide technique in the 8–12 GHz frequency band.



(a)



(b)

FIG. 4. (a) Rectangular waveguide method: Permittivity spectra of a KTN thin film deposited on MgO substrate and (b) Cavity perturbation method: Transmission coefficients (modulus) of the cavity loaded with an MgO substrate and with a KTN/MgO bilayer.

The dielectric properties of the ferroelectric film appear to be non-dispersive in this band, with mean values of 343 and 32.6 for the real and imaginary parts of the permittivity, respectively.

B. Cavity perturbation method

The second method, developed at Lab-STICC, enabled us to extract the dielectric properties of the ferroelectric thin film from the variations in resonance frequency and the quality factor of a rectangular cavity induced by the insertion of the material under test. The resonant cavity consists of a 5-cm long X-band rectangular waveguide section excited by circular apertures. For this structure, the resonant modes are of $TE_{10\delta}$ type. When the integer number δ is odd, the electric field shows a maximum magnitude at the centre of the cavity. The sample to be characterized was placed in this region to increase the sensitivity of the method. A slot, situated at the centre of the cavity, enabled insertion of the sample.

Two steps were then required to characterize the ferroelectric thin film. First, an MgO substrate was inserted into the cavity. The resonance frequency and the quality factor of the cavity were calculated and were considered as reference values. Second, a KTN/MgO sample was inserted into the cavity. At this step, the MgO substrate must present the same dimensions and dielectric properties as the one considered in the first step. Thus, the dielectric constant and losses of the KTN thin film modify the resonance frequency and quality factor of the cavity. As variations of these parameters are low, the complex permittivity of the ferroelectric thin film can be calculated analytically using a low perturbation approach.^{32,33}

In our case, the cavity loaded with the MgO substrate showed a resonance frequency of 6.7403 GHz and a quality factor of 3768 for the TE_{101} mode. When a KTN/MgO sample was inserted into the cavity, the resonance frequency shifted to 6.7287 GHz and the quality factor decreased to 1839 (Fig. 4(b)). The low perturbation approach then enabled the dielectric properties of the thin film to be extracted: $\epsilon' = 312$ and $\tan\delta = 8 \times 10^{-2}$.

C. Resonant method using a cylindrical cavity and dielectric resonators

XLIM developed a resonant system at 12 GHz, made up of a cylindrical cavity associated with two identical dielectric resonators placed face to face inside the cavity (Fig. 5).³⁴ The variations of resonance frequency and the quality factor of the $TE_{01\delta}$ mode induced by the insertion of a sample (substrate, thin film + substrate) between the dielectric resonators allows its permittivity and loss tangent to be extracted using simulation software based on the method of lines (MOL).

The dielectric properties extracted using this method are extremely sensitive to the accuracy of information on the thicknesses of the different materials, which is notably difficult to measure accurately in ferroelectric thin films. New methods are currently being sought to minimize this measurement uncertainty. Another major limitation of this method concerns the permittivity-thickness product, which

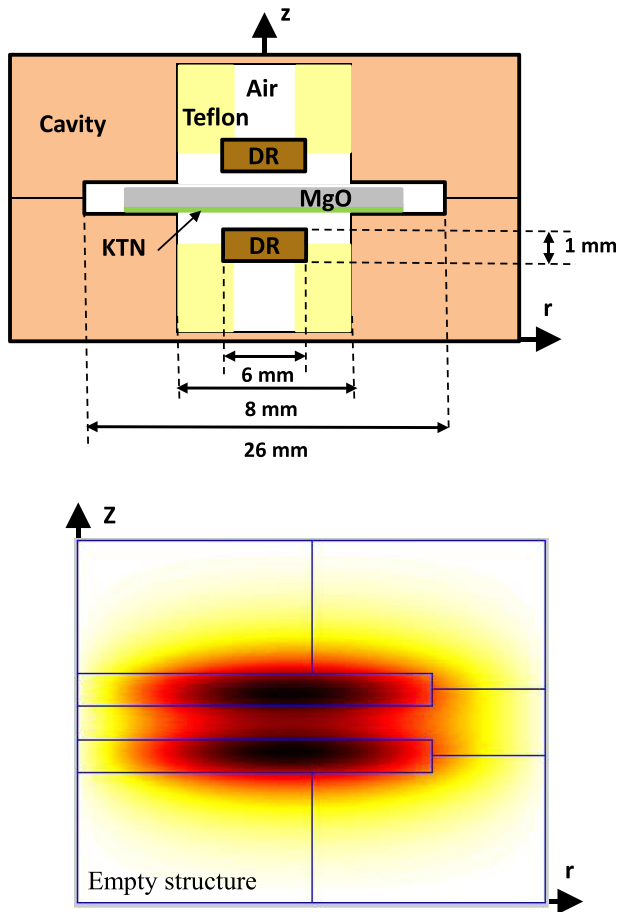


FIG. 5. Measurement cell and electric field in the structure for the three measurement steps.

must be sufficiently significant to induce noticeable changes between the calibration (substrate) and measurement steps (thin film + substrate). A rigorous EM analysis is then needed to extract the dielectric properties of the thin film. Our analysis was based on the 2.5D MOL, which allowed us to take into account very thin dielectric layers and avoid meshing problems associated with 3D methods. The complexity of the structure required that a successive approach be used for the extraction of the dielectric properties, whose convergence is ensured by the Newton-Raphson method.

D. Broadband characterization using a CPW structure

A broadband characterization method (40 MHz–67 GHz) based on the measurement of the lineic electric parameters (capacity C and conductance G) of coplanar transmission lines (CPW) was used at IMEP-LAHC. Following this method, the KTN layer to be tested was first deposited on an MgO substrate, then metallic electrodes were deposited on the KTN before chemical engraving (Fig. 6). Metallization dimensions were chosen by making a compromise between maximizing the interaction between the electric propagated field and the KTN layer, and minimizing conduction losses in the CPW line. S-parameters of the CPW line were measured using an Agilent network analyzer (PNA-XN5247A) calibrated by an LRRM procedure and treated to obtain the values of C and G . Extraction of the

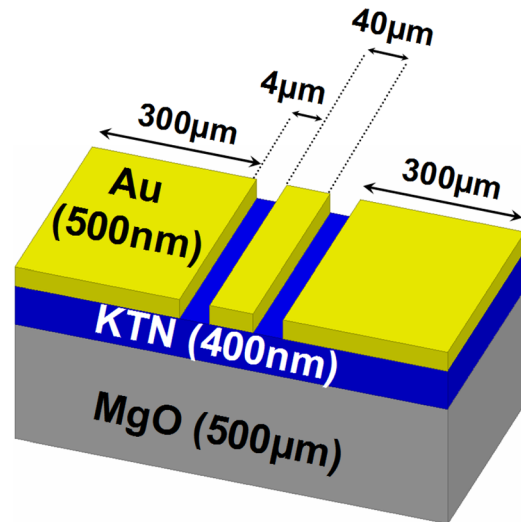


FIG. 6. Cross-section of the CPW structure.

complex permittivity of the KTN thin film was then performed through an iterative optimization process based on Powell-Simplex algorithms.³⁵ Real and imaginary parts of the estimated thin film permittivity were adjusted simultaneously until the values of C and G simulated by the EM solver HF2D fit the measured values.³⁶ To do this, a specific error function, strongly dependent on the permittivity value and defined in a two-dimensional space (ϵ' , ϵ''), was minimized. This extraction process is used at each frequency and offers the benefit of covering a very wide range of frequencies. This reveals a distributed relaxation of the KTN permittivity, which is well described by a Cole-Davidson model. Fig. 7(a) shows the good agreement between measured and calculated data. The choice of this model is well supported in the highest part of the frequency band by the pronounced decrease of the real part and the increase of the imaginary part of the permittivity.

The coplanar lines deposited on KTN and characterized in IMEP-LAHC were also measured at Lab-STICC to retrieve complex permittivity using a different data processing program. Here, the theoretical analysis based on quasi-static approximation consists in using only the transmission

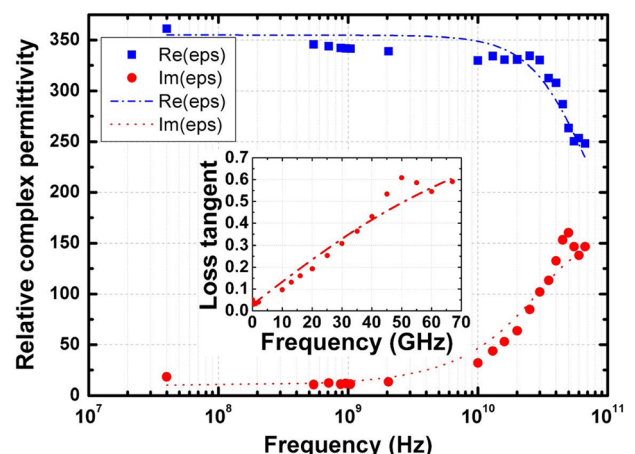


FIG. 7. Measured (symbol) and calculated (line) complex permittivity spectra of the KTN thin film. Insert: dielectric loss tangent.

parameter S_{21} in the case of non-magnetic materials. The permittivity of the thin film was calculated from the measured effective permittivity of the structure. Using the conformal mapping technique, we obtained analytical expressions that related the complex permittivity of the thin film to the geometrical parameters of the line and the dielectric properties of the substrate.³⁷ These analytical relationships enabled us to avoid using numerical optimization procedures to retrieve thin film permittivity. In this method, the different types of losses in the CPW structure were also taken into account: we considered the substrate dielectric, conductor, and radiation losses.²⁵

The CPW lines were measured using the Cascade Microtech[®] probe station and Agilent E8364 VNA in the frequency range [5 GHz–50 GHz]. During the first stage, the device was calibrated using an LRRM procedure. We then performed a differential measurement on two lines of different lengths in order to minimize contact inaccuracy. The measured complex permittivity and loss tangent are shown in Fig. 7.

V. COMPARISON AND DISCUSSION OF EXPERIMENTAL RESULTS

The choice of characterization methods for this inter-comparison was dictated by their complementarities. First of all, we wanted to compare the suitability of the two usual operating modes for the characterization of thin films; on the one hand, resonant techniques (cavities, resonators) and, on the other hand, the reflection/transmission methods (transmission line, waveguide). For broadband techniques, the interest of our study lies in the comparison between a differential approach (correction of S-parameters by taking into account the measurement of the empty cell) and an overall measurement in CPW line, in which the thin film is integrated. It would be also interesting to study the influence of the theoretical models used for the EM analysis of the different measurement cells on the accuracy of the measured data.

A. Influence of the models used for the EM analysis of the cell

The first broadband approach, based on the use of a rectangular waveguide, can be considered as non-invasive because of the absence of metallization onto the ferroelectric film, in contrast to the second, for which the CPW metallic layers were directly deposited on the thin-film. As discussed in Sec. II, the latter method does not allow removal of cell imperfections in the absence of a sample (empty cell). In these conditions, the only alternative for the data processing procedure is to accurately predict all the physical phenomena inherent in the measurement cell: skin depth effect in the metallic layers (central strip conductor and ground planes), radiation, dispersion of the EM properties of the substrate, etc. The problem with this approach is the validity of the theoretical model used for the EM analysis of the CPW line. The model must thoroughly describes the skin depth effect in metallic layers without being limited by the high contrast between the respective thicknesses of the different layers present in the structure: a few hundred nanometers for the

ferroelectric thin film and a few hundred micrometers for the substrate. To bypass this theoretical difficulty, IMEP-LAHC chose to exploit the lineic parameter capacity C and conductance G of the CPW line, which are not strictly dependent on conductor loss. The modeling of these two quantities was done using an EM solver, which ensures very good calculation accuracy. The difficulty related to the choice of the model for EM analysis of the measurement cell can be illustrated by a comparison of the loss tangent spectra retrieved from the two different data processing programs used with the CPW line. In the [5 GHz–50 GHz] frequency band, the quasi-static approach gives a variation in loss tangent from 0.12 up to 0.25 (Fig. 8(b)), whereas the full-wave analysis predicts variation from 0.04 up to 0.6 (Fig. 7(b)). Since the full-wave analysis enabled us to determine measurable parameters that were independent of conductor loss (capacity C and conductance G), we can conclude from this comparison that the loss models related to the quasi-static approach do not allow accurate values to be retrieved for the dielectric loss of the ferroelectric thin film. The real parts of the permittivity spectra obtained from the two different programs used to process the CPW measured data were no longer in very good agreement in the upper part of the exploited frequency range (around 33% relative discrepancy at 50 GHz, see Figs. 7 and 8).

For the rectangular waveguide method (see Sec. IV A), a full-wave EM analysis was also necessary to guarantee high accuracy of the calculated S-parameters. In addition to the

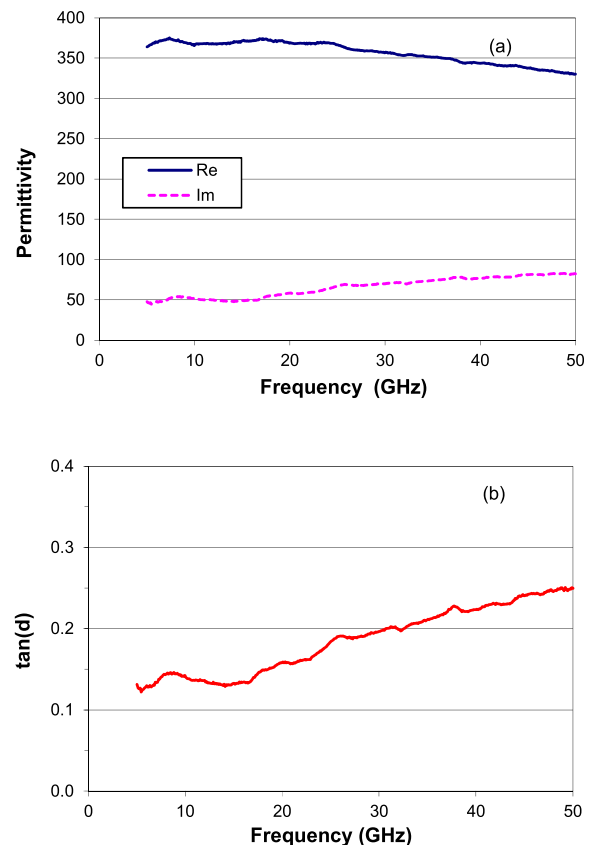


FIG. 8. CPW structure-based characterization of KTN thin film deposited on MgO: (a) complex permittivity spectra, (b) dielectric loss tangent.

TE₁₀ dominant mode, six higher-order modes were taken into account in the EM analysis to ensure convergence of the calculations. It is not necessary to have a very accurate description of the skin effect in the metallic walls of the waveguide because of the differential character of the measurement procedure (calibration of the S-parameters by removing the empty cell measurement). Since the data processing programs used to retrieve the permittivity of the ferroelectric thin films from the rectangular waveguide and the CPW line measurements were both based on a full-wave analysis, it is interesting to compare the corresponding results. When comparing the permittivity spectra in Figs. 4(a) and 7(a), we observed a very good agreement between the measured data obtained from both broadband methods based on full-wave analysis. At 10 GHz, the real part of the permittivity is equal to 340 and the loss tangent is equal to 0.09 for both methods. These data are in very good agreement with those obtained from the resonant method based on a full-wave analysis ($\epsilon_r' = 352$, $\tan\delta = 0.11$ at 12 GHz). In contrast, the low-perturbation approach used with the rectangular cavity predicted a lower value for the real part of the permittivity ($\epsilon_r' = 312$ at 6.7 GHz).

From these comparisons, we can conclude that it is preferable to use full-wave analysis in the description of the microwave behavior of measurement cells as this guarantees a higher accuracy of measurements of complex permittivity of ferroelectric thin films.

B. Error analysis

Results from the measurement campaign on KTN thin films are summarized in Table I. Despite the opposing characters of the operating modes (resonant versus Reflection/transmission techniques) and, in the case of broadband methods, despite the different propagation structures (waveguide versus transmission line) and the different systematic error correction procedures (differential measurement versus global measurement), there is good agreement between dielectric properties retrieved from full-wave analysis-based data processing programs.

The resonant method that uses a cylindrical cavity offers the least sources of uncertainty. This method is based on a differential-type correction of systematic errors, in which the quality factor and the resonance frequency of the empty cavity constitutes the reference. Cavity modeling is rigorous,

and the existence of an air gap is not an obstacle. Moreover, measured parameters are less sensitive to random errors (VNA signal to noise ratio) than broadband techniques. However, permittivity is determined at a specified frequency, so an assessment of the dispersion of dielectric properties in ferroelectric thin films is not possible. The main source of uncertainty is related to inaccuracies in the determination of layer thickness due to its inhomogeneity. For the KTN ferroelectric layer characterized during these comparison measurements, it should be noted that the Pulsed Laser Deposition technique used for its construction ensures a relative good homogeneity of layer thickness over the whole surface. The mean relative variation observed for the thickness of submicronic ferroelectric thin films deposited by PLD is around 15%³⁸ over the whole tested surface. This level of inaccuracy leads to a relative error of 12% for the retrieved permittivity (real part) of the layer. This is confirmed by a previous experimental study,¹³ which showed that it is possible to measure real permittivity of ferroelectric films having submicronic thickness and permittivity range of 100–10000 with uncertainties similar to uncertainties about their thickness. Finally, in this campaign, permittivity measured in the cavity can be considered as the reference value fixed here at 12 GHz.

A resonant method based on a rectangular cavity was also tested. Using this approach, permittivity was calculated by the approximation of small perturbations. Even though the resonant frequencies of the two cavities were not identical (12 GHz and 6.73 GHz), their relative proximity led to a difference of 11% for the real part of the permittivity and 28% for the loss tangent. This comparison made it possible to estimate the uncertainties brought about by using the weak perturbation assumption to predict the EM field pattern variations in resonant cavities when inserting a dielectric sample.

The two broadband methods led to comparable values of permittivity, close to the reference value obtained from the cylindrical cavity-based method: $\epsilon_r = 352$; $\tan\delta = 1.12 \times 10^{-1}$. Compared to these reference values, the differences observed at 12 GHz between the waveguide-based method and the method using the CPW structure were 2.5% and 6.2%, respectively, for the real part of the permittivity and 15% and 10% for the loss tangent.

The error source analysis related to the rectangular waveguide-based broadband method showed that the main uncertainty in the measured complex permittivity is due to

TABLE I. Results overview of KTN (500 nm) thin film deposited on MgO (500 μm) substrate.

EM structure	Destructive character	EM analysis of the cell	Operating mode	Frequencies (GHz)	ϵ_r	$\tan\delta$
Rectangular waveguide ^(a)	Non invasive	Full-wave	Broadband	8–12	343	9.5×10^{-2}
Rectangular Cavity ^(a)	Non invasive	Quasi-static	Mono-frequency	6.73	312	8.0×10^{-2}
Resonant System (cavity + DR) ^(b)	Non invasive	Full-wave	Mono-frequency	12	352	1.12×10^{-1}
CPW line ^(c)	Invasive	Full-wave	Broadband	0.04–67	341 (1 GHz) 248 (67 GHz)	0.033 (1 GHz) 0.59 (67 GHz)
CPW line ^(a)	Invasive	Quasi-static	Broadband	5–50	365 (5 GHz) 330 (50 GHz)	0.12 (5 GHz) 0.25 (50 GHz)

^(a)Lab-STICC

^(b)XLIM

^(c)IMEP-LAHC.

imprecision on the permittivity value of the substrate of the thin film.³⁰ This error occurred in all broadband methods used for this campaign. However, its influence can vary according to the measurement cell and how the EM wave interacts with the substrate. For example, the CPW structure minimizes this influence because the EM energy is strongly concentrated between the central conductor and the ground planes, around the interface separating the air and the ferroelectric thin film.

Existence of air gap between the bilayered sample and the walls of the waveguide can lead to an underestimation of the real part of the permittivity. Particular attention must be given during the machining of samples in order to minimize uncertainties due to the air gap. This does not occur with the CPW-based method. An error analysis performed during the use of the rectangular waveguide method³⁰ enabled us to estimate the influence of an air gap on the accuracy of the measured permittivity. We predicted a relative uncertainty of 10% for the real part of the permittivity (value around 350) for a 10- μm thick air gap. The imaginary part of the permittivity is slightly sensitive to air gaps. A one-centimetre length slot could be machined in the median plane of the rectangular waveguide to facilitate the insertion of the sample to be characterized. This new way of inserting the sample could also make it possible to eliminate the error caused by air gaps.

VI. CONCLUSION

The unique character of ferroelectric thin films, which results from their submicron thicknesses and related high permittivity values and moderate losses, makes their microwave characterization difficult. This difficulty is increased by the impossibility of separating the thin layer from its substrate. The large difference between the thicknesses of substrate and thin layer requires an accurate determination of the latter's EM properties. Fortunately, this geometric contrast is balanced by a difference in permittivity values in favor of the thin layer.

The first lesson that should be drawn from these inter-comparison permittivity measurements performed on a KTN thin layer, is the need to choose thorough theoretical approaches for the EM analysis of the measurement cells. The full-wave analysis (mode-matching method, finite elements method, etc.), which takes into account the physical phenomena inherent to the structures in which the thin layer is inserted (or directly integrated) for characterization, must be consistently favored over quasi-static theories (weak perturbation approximation, quasi-TEM assumption, etc.). This conclusion is logical since the contribution of the thin layer to the measured S-parameters is always in the minority compared with the contributions of the other elements, particularly the metallic layers and substrate.

The most surprising conclusion drawn from the results obtained in this study is that the differences observed between the loss tangent determined from broadband methods and that given by the resonant technique are relatively low. The greater sensitivity of the broadband methods to the signal-to-noise ratio could suggest a far greater difference than that actually observed. This good result demonstrates the relevance of the

two broadband methods tested for the EM characterization of ferroelectric thin layers. The waveguide-based method could be defined as a standard for EM characterization of dielectric thin films as long as the data processing program and related experimental protocol allow: (1) elimination of the air gaps (or correction according to their influence) and, (2) use of the measurement performed on the substrate of the thin layer as a reference in the differential approach. The CPW-based method is essential for *in-situ* characterization over a broad range of frequencies. It also offers the advantage of allowing the study of the variation of ferroelectric layer permittivity under the action of a static field.

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